Fiber-integrated nano-optical antennas and axicons as ultra-compact all-fiber platforms for luminescence detection and imaging down to single nano-emitters

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Thèse présentée par

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pour obtenir le
Grade de Docteur de
l’Université de Franche-Comté

Spécialité : Optics

Fiber-integrated nano-optical antennas and axicons as ultra-compact all-fiber platforms for luminescence detection and imaging down to single nano-emitters

Soutenue publiquement le 05 July 2016 devant le Jury composé de :

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ACKNOWLEDGEMENT

First and foremost, I would like to express my sincere gratitude to my supervisor Mr. Thierry Grosjean, the researcher of CNRS, for his continuous support and help during my PHD study with great patience, motivation and knowledge. He offered me a lot of useful advices and guidance in theoretical and experimental researches, and also in this thesis writing. He encouraged me to be an independent researcher and gave me great confidence to finish this challenging subject. I learned a lot during the last three years in knowledge and how to become a good researcher with the help of Thierry. I also want to acknowledge my cosupervisor Mr. Jean-Marc Merolla. During the last three years, he taught me how to perform an experiment in a rigorous way. I appreciate all their contributions of time, ideas and fundings to make my PHD finish successfully.

Besides my supervisors, I would like to thank my thesis manuscript reviewers: Mr. Ulrich Fischer and Prof. Didier Tonneau. Many thanks to them for their careful corrections of my manuscript and for their valuable questions during my defense, which widened my knowledge and research interest. Many thanks to them also for their recommendation and approval of my research of my PHD, which encourages me to continue research career in my future life. Thanks Mr. Johanne Cussey and Herve Maillotte for honouring me with their presence to my PHD thesis defense.

I would also like to acknowledge the engineers in FEMTO-ST, MIMENTO and Lovalite for their help in the experiments preparations. Thanks engineer Miguel who helped me a lot in experiments and defense preparation. Thanks Roland for antenna fabrication with FIB. Thanks Dusan for his patient instruction for fiber tip fabrications. Also thanks Laurent for his help in quantum dot samples preparation. I also learned a lot from them when I work together with them.

Also many thanks to the friends I met in FEMTO-ST for their help and accompany during the last three years. Thanks Dr. Huihui Lu who helped me a lot in life when I just arrived France. Thanks my friends, Abdoulaye, Shanti, Yannick, Ali, Eli, Xavier, Alexis, Nyha, Taseen, Tania, Venacio, etc... I would also like to thank my Chinese friends I met in France, Xie Chen, Qi Wei, Guoping, Xu Xin, Wu Chang, Mengjia, Ma Chen, Wang Yanfeng, etc... We had many happy times in France and it will always be very precious memory in my life. Thank you for helping me a lot when I had difficulties. Special thanks to Wendy, who came to France together with me. It's lucky that we two finished the PHD successfully at the end. All the difficult times we experienced together make us stronger and more mature.

At last but not least, I would like to acknowledge my family, my parents, my parents-in-law, my brother Hongxu, my sister-in-law Yang Leilei and my little nephew Tiange, who supported and encouraged me to leave China and come to France for the PHD research. Also special gratitude to my husband Feng Cong who is in Hongkong for PHD research. It's a great challenge for us to be so far away from each other for so long time. Thank you very much for your understanding and encouraging me at the most difficult times. I gained a lot during the three years. We both are getting more mature.
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Luminescence is commonly defined as light emission by matter and is one of the fundamental result of light/matter interactions [1]. To produce light, matter generally needs to absorb external energy beyond its intrinsic energy band-gap responsible for light emission [2]. Different excitation energies are usually applied to matter for achieving luminescence phenomenon, for instance, UV light which leads to fluorescence and phosphorescence phenomena [3], high energy radiations (X-rays, Gamma rays, etc) [4, 5], electricity which leads to electro-luminescence [6], free space launched electrons which originates cathodo-luminescence [7]. Luminescent materials and techniques have taken in the past decades a crucial role in many scientific, medical and industrial domains of high societal and economical perspectives, thus strongly impacting our daily life as shown in Figure 1. As an example, in medical diagnostic systems, X-ray luminescence computed tomography (XLCT) has been developed to initiate tumor diagnosis by exploiting X-ray excited luminescent materials [8]. Electroluminescence is widely applied in display panels and TV screens. It is also widely used in lamp engineering [9], which is also the case for fluorescence. In biology research, fluorescent labels are an appealing choice for tissue and organ imaging [10]. With the development of nanotechnologies, the concept of single photon sources control arose for a large panel of applications in telecommunications and cryptography [11]. Luminescent materials can also be found in many other applications such as solar cells engineering [12], oxygen sensing [13], etc.

**Figure 1:** The daily-life applications of luminescent materials.

Luminescence detection is often a central process in many applications, especially in 1
biomedical diagnose and imaging, medical and industrial sensing and quantum information processing. Spherical optics (objectives, lenses) is of prime importance to achieve luminescence collection and it has been used for many applications. Last generations of X-ray cameras are based on luminescent screens and use microscope lens to couple the luminescent material to a CCD camera [14] to improve imaging resolution. Conventional far-field microscopies, as well as confocal microscopy, are based on spherical optics [15]. These techniques take a central position in luminescence imaging and characterization down to the wavelength scale [16]. For instance, confocal microscopy has been success- fully applied in molecular imaging with subcellular resolution, enabling the visualization of morphological details in tissues [17]. Far-field scanning microscopy was used to image in vivo tissues with X-ray excitation [18] and also small scale electroluminescent systems [19]. Conical optics (axicons [20]) is another attractive solution to detect luminescence. The outstanding long focus depth of axicons has been successfully applied in fluorescence imaging of 3D thick sample with high perspectives in medical inspections [21].

These microscopy systems suffer however from important limitations. First, they often use bulky optics and thus their compactness and flexibility are limited. They are for instance not compatible with endoscopy and sometime highly space consuming. Their out-of-lab implementation can be also expensive. Moreover, multi optical element benches are often unavoidable, which may require strict alignment processes that are tricky for end-users (non specialists in optics). The development of low-cost plug-and-plays systems in the domain of luminescence collection and imaging is thus highly desirable. Spherical optics also show limited chromatism and are thus not well-adapted to infrared fluorescence imaging, especially with visible excitation of fluorophores. Finally, far-field luminescence collection techniques is limited by diffraction to half wavelength resolution.

Optical fibers offer an interesting alternative for luminescence detection with the advantages of being compact, flexible and low-cost. With optical fibers, one can also expect the achievement of self-aligned plug-and-play probes and sensors. Optical fibers have been already applied for luminescence collection in frame of sensor technology. Oxygen sensors rely on in-fiber fluorescence collection, the fiber end is coated by oxygen sensitive luminescent materials, whose luminescence is known to be quenched by oxygen [22]. Another important application of optical fibers is luminescence endoscopy, which is used for instance to monitor radioactive labels in patients (Cerenkov luminescence endoscopy) [23]. Besides medicine, biology and chemistry, quantum information processing could also take benefit of in-fiber luminescent collection. On-demand single photon sources (i.e. fluorescent quantum emitters) and single photon detection take central position in this domain. Single photon sources, such as quantum dots (QD), fluorescent molecules, diamond color centers have been developed. The development of fiber on-demand single photon sources would be very desirable as it would open the perspective of single photon sources in fiber networks and would thus unlock the full potential of these emitters in many different scientific and industrial domains of high societal and economical impact. Several research groups are starting to move towards this direction.

My thesis is devoted to develop ultra-compact, plug-and-play and low-cost fiber optical systems for in-fiber luminescence collection. In other words, the goal is to imagine and develop new devices for efficiently optically coupling fluorescent emitters to single-mode fibers. Targeted applications are here fluorescence microscopy, sensing and fibered single photon emission. First, a new far-field in-fiber fluorescence coupling will be proposed and demonstrated with the use of non-diffracting Bessel beams, thanks to an ultra-compact and fiber self-aligned axicon. Such beams will be shown to provide the first
resolved infrared fluorescence imaging of PbS QDs, where spherical optics fails due to chromatism problems. Then, in-fiber near-field coupling will be achieved by means of the concept of double resonance bowtie nanoaperture antenna (BNA), with demonstration of nanometer resolution all-fiber near-field imaging of single PbS (infrared) QDs. Such fiber-integrated near-field microscope is ultra-compact since it avoids the use of bulky optics: fluorescence excitation and collection is achieved locally down to the nanometer scale through the same fiber. Finally, the concept of fiber nano-optical horn antenna will be proposed and investigated for in-fiber luminescence out-coupling. First investigation will concern the generation of fiber-integrated single photon sources. The concept will also be demonstrated in the coupling of X-ray excited luminescence into single-mode fibers, for generating the first ultra-compact fiber-integrated X-ray sensors and real-time dosimeters with high perspectives in radiotherapy real-time control. We explore here for the first time to our knowledge the concept of nano-optical antenna to control X-ray excited luminescence (here directionality). Generally, nano-optical antennas are "limited" to the control of fluorescent emitter, not yet X-ray sensitive scintillators.

This manuscript is divided in 4 chapters.

In a first chapter, I will introduce the state of the art relative to this work. It concerns mainly fluorescence excitation, collection and control. Most of the introduced techniques deal with optical microscopy and fiber detection. Confocal and conventional far-field scanning microscopies and axicon systems will be detailed in luminescence detection and imaging. The role of nano-optical antennas in fluorescence control will then be shown. Then, direct coupling between single fluorescent emitters and optical fibers, which avoids the use of conventional bulky optics, will be presented as well as the essential role of nano-optical antennas in that domain.

In the second chapter, I will introduce and demonstrate the concept of AXIGRIN (Axicon on GRIN lens), as a two-way far-field communication channel between fluorescent emitters and an optical fiber, for ultra-high depth of field fluorescence imaging. This compact component turns fiber output waves into a non diffracting Bessel beam, and reciprocally it couples fluorescence in 3D deep sample into a fiber without the need of tomographic techniques. Resolved mappings of infrared fluorescence from PbS colloidal QDs have been demonstrated for the first time to our knowledge onto patterned test-objects. Since the AXIGRIN is self aligned with the optical fiber, no more complex optical alignment is needed, which makes the system compact, flexible and easy to use.

In a third chapter, I will present and demonstrate the concept of double resonance BNA for probing single quantum emitters in a all-fiber SNOM (scanning Near-field Opticam Microscopy) set-up that does not need bulky optics. This "achromatic" fiber SNOM nanoprobe allows for both locally exciting and collecting (down to the nanometer scale) fluorescence from single point-like emitters. Two resonances, centered to the absorption and emission wavelengths of the emitters, can be tuned independently from each other (depending on practical requirements), by modulating the nano-antenna geometrical parameters and size. In the frame of this study, they will be suitable for single PbS QDs imaging, showing very separated absorption and emission wavelengths, 808 nm and 1500 nm, respectively. Such a spectral mismatch between excitation and emission in a fluorescent process is far beyond chromatism of spherical optics. The first SNOM images of these QDs validates the proposed concepts for all fluorescent emitters.

In the fourth and last chapter, the concept of fiber nano-optical horn antenna will be
proposed and investigated for in-fiber luminescence out-coupling. First investigation will concern the generation of fiber-integrated single photon sources. The concept will also be demonstrated in the coupling of X-ray excited luminescence into single-mode fibers, for generating the first ultra-compact fiber-integrated X-ray sensors and real-time dosimeters with high perspectives in radiotherapy real-time control. We explore here for the first time to our knowledge the concept of nano-optical antenna to control X-ray excited luminescence (here directionality). Generally, nano-optical antennas are "limited" to the control of fluorescent emitter, not yet X-ray sensitive scintillators.

The manuscript will be ended with a general conclusion.
STATE OF THE ART FOR LUMINESCENCE DETECTION

The techniques for luminescence detection and imaging will be reviewed in this chapter. Refractive lens and optical fibers are common techniques in luminescence detection which will be presented in the first two sections. Nanoantenna will then be introduced as a powerful solution to tailor fluorescence emission from single emitters, in terms of fluorescence signal and directionality enhancements. Nano-antennas are thus clearly in favor of luminescence detection improvement. The union of a nano-antenna and a single emitter can be seen as a “super emitter”. The chapter will be divided into three sections:

1. Refractive lens system for luminescence detection. The applications in confocal microscopy, scanning far-field and near-field microscopies, involving spherical and conical optics for luminescence excitation and detection will be reviewed.

2. Optical fiber system for luminescence detection. "Fiberscope" application in imaging tissues in vivo, fiber system application in X-ray excited luminescence detection and various developed fiber systems for single photon detection will be included in this section.

3. Nanoantenna-assisted super emitters. The role of nano-antennas in tailoring (enhancing) fluorescence absorption and emission (in order to enhance fluorescence detection), will be reviewed.

1.1/ Refractive lenses for luminescence detection

Optical microscopy is a basic, simple and widely used technique in luminescence detection and imaging. Confocal microscopy is another technique for luminescence detection and imaging with the advantages of slightly higher resolution. It has been successfully applied in fluorescence sensing and labeling of proteins in living cells [24]. In microscopy systems, objective (i.e. magnifying high numerical aperture combination of spherical lenses) is a critical element for luminescence detection and imaging. It decides the angle range of collected light and consequently the imaging lateral resolution. Especially in single photon detection, high numerical aperture lens is needed to collect maximum optical signal from fluorescent single photon sources. It is necessary to develop new kinds of objectives with higher numerical aperture. Oil-immersion objective has been applied in
microscopies with numerical aperture larger than 1 and were successful in imaging fluorescent elements down to single molecules [25, 26]. Except oil-immersion lens, solid immersion lens is another choice to enhance even more the numerical aperture of microscopy systems.

The concept of solid immersion lens originates from oil-immersion lens. It is an optically transparent hemisphere lens with high refractive index. Except for their applications in lithography, high-density data storage, imaging, etc, solid immersion lens-based confocal microscopy has also been exploited to improve the single photon detection efficiency. The high refractive index of solid immersion lens increases the numerical aperture of the microscopy system to unprecedented values. The most straightforward consequence is that the fluorescence in large emission angle can be collected by the microscopy system. Conversely, the excitation light can be focused onto very small area with increased intensities. Since the excitation rate of quantum emitters is proportional to the excitation intensity, it is also improved. In addition, it has been shown theoretically that when an emitter is put at the center of a solid immersion lens flat surface, its emission pattern is also modulated due to large refractive index difference between solid immersion objective and air [27] and more photons are emitted toward the solid immersion lens as shown in Figure 1.1(a). The modulated emission pattern also contributes to collection efficiency enhancement. The combination of enhanced excitation rate, modulated emission pattern and large numerical aperture of the objective, leads to greatly enhanced detected signal from single photon source.

![Solid Immersion Lens Diagram](image)

**Figure 1.1:** Scheme of solid immersion lens concept for fluorescence excitation and detection. (a) Scheme of a solid immersion lens concept, with modified emission diagram of a dipolar point-like emitter [27]. (b) Experiment setup of solid immersion lens in application of single NE fluorescence collection [28].

Siyushev’s group fabricated a micro solid immersion lens which is made of diamond [28]. The diamond is the host material for single color centers and a native single NV defect was formed at the center of the solid immersion lens flat surface. The NV center was excited by the laser of 532 nm and emit photon in broadband wavelength (the character-
istic zero-phonon line wavelength of 637 nm). A 0.85 numerical aperture objective was employed to collect the fluorescence. Based on the experimental setup as shown in Figure 1.1(b), the detection efficiency was enhanced by sixfold as compared to the detection efficiency of only the same conventional objectives with numerical aperture of 0.85. Aberration exists in this case and due to this, the experimental enhancement is slightly smaller than theoretical predictions.

Generally, scanning optical microscopy and especially confocal microscopy are based on spherical optics for which chromatism is unavoidable. There is a nondiffractive conical optical element—axicon, which can also be used in luminescence imaging with long working distance.

Axicon is a conical lens, which can ideally transform a point-like source on axis to a needle of light of extended length [20]. It can also change a collimated beam into a non-diffracting Bessel beam [29]. Due to the property of long focal depth, axicon can be used to image a 3D thick sample, such as brain tissue, with all the 3D information collected and recorded in a 2D image. It has been successfully applied in two photon fluorescence microscopy to image 3D thick specimens [21, 30, 31].

A BK7 axicon (1 cm diameter, angle $\alpha$ of 30° and 6.8 mm thickness at the center) has been experimentally applied for luminescence imaging [21]. In the scheme in Figure 1.2(a), a femtosecond Ti:sapphire laser source spectrally centered at 800 nm and of average power of 300 mW was used. A sample of AGAR-10% with the thickness of 1 mm was prepared by dispersing 15 µm diameter fluorescent microspheres (Triton Technology, Fluorescent Lemon) throughout the sample. Bessel beam which is composed of a bright
central spot and several concentric rings was generated by the axicon. The central spot FWHM size was about 1µm and the field depth was 2.4 mm. Due to the large field depth of the Bessel beam, the luminescent microspheres throughout the sample thickness can be excited by the Bessel beam evenly. Since most intensity of the Bessel beam is focused onto the central part, the excitation of the concentric rings can be ignored. The fluorescence from the sample was also collected by the axicon. By the reflection of a dichroic splitter, the collected signal was separated from the excitation light and redirected to a photomultiplier tube at end. With the large field depth, the optical signal of the evenly excited microspheres throughout the sample can be detected by the axicon at the same time. For conventional objective imaging, the excitation light was focused onto a single surface and only the microspheres right at that surface can be imaged. The comparison of the scanning results from the two imaging methods is shown in Figure 1.2(b) and (c), respectively. The sum of 12 objective scanning images is shown in (b), but with the axicon, only single scanning is enough as shown in (c). So compared to traditional objective imaging, the axicon imaging is obviously more efficient and rapid imaging 3D samples.

Spherical optics, and more especially objectives, are also included into SNOM architectures for collecting the fluorescence signal from nanoprobes. It has been for instance used to detect the fluorescence photons from Eu³⁺-doped nanocrystals [32] used as nanoprobes for revealing new optical information onto samples. A nanocrystal is glued at the apex of a near-field scanning optical microscope (SNOM) tip and controlled by the SNOM system to raster scan sample surface. The experimental setup is shown in Figure 1.3(a), where the nanocrystal was excited by oblique incident light at 532 nm. The image is formed during the raster scan by collecting the fluorescence signal with a 0.8-NA objective coupled to a spectrometer and cooled CCD camera.

Figure 1.3: (a) Experimental setup. The inset is scanning electron microscope (SEM) image of Eu³⁺-doped nanocrystal (200 nm diameter) which was attached onto tungsten SNOM tip. (b) Luminescence spectra of the nanocrystal at several distances from a gold mirror. (c) Scanning images in x-y plane. The images from left to right: topography of the gold stripe; magnetic dipole luminescence image and two electric dipole luminescence images. The image size is 3×4.8µm². (d) The scanning images in y-z plane. Image size is 4×1µm² [32].
1.2. FIBER SYSTEM FOR LUMINESCENCE DETECTION

The typical spectrum of \( \text{Er}^{3+} \)-doped nanocrystal, displayed in 1.3(b), shows three dominant peaks, which correspond to one magnetic dipole (MD) and two electric dipole (ED) luminescence emissions. By detecting selectively (with the objective and proper filters) the fluorescence from the electric and magnetic dipole emissions, the authors could plot separately the electric and magnetic optical images right at the sample surface. Figure 1.3(c) represents images obtained by collecting the fluorescence from the nanotip during raster scan in contact to a gold stripe. Topography, magnetic and electric dipole luminescence mappings are arranged from left to right. The scanning images in longitudinal plane are shown in Figure 1.3(d). The magnetic and electric dipole luminescence images arranged from left to right.

Spherical optics have also been applied in the collection and imaging of X-ray excited luminescence [33]. In the experiment shown in Figure 1.4, \( \text{Gd}_2\text{O}_3 \) sample was excited by X-ray and the luminescence was collected and collimated by a fused quartz lens. After the mirror reflection, the luminescence was focused by another lens onto an air-cooled photomultiplier (PMT). Thus the optical lenses system were successfully applied in X-ray excited luminescence detection.

Figure 1.4: The experimental setup of X-ray excited luminescence detection [33].

Although spherical and conical optics are successfully applied in fluorescence collection and imaging, they still undergo strong limitations given their “bulky” nature. Moreover, the strict alignment requirements of the multi-element optical systems limit the end-users to those who are expert at optics. Finally, refractive optics suffer from chromatism that limits their use to visible luminescence imaging. Infrared luminescence imaging remains difficult despite promising applications. Due to these limitations, it would be very attractive to find an alternative which is more flexible, easy to control and low-cost, to achieve luminescence detection.

1.2/ FIBER SYSTEM FOR LUMINESCENCE DETECTION

Fiber system is an alternative to achieve efficient fluorescence detection. It has been applied successfully in bio imaging [34], sensing [35], etc.

Fiber-based two-photon fiberscopy has been experimentally demonstrated for in vivo
functional imaging in living mouse brain [36]. The experimental setup is shown in Figure 1.5(a). The device consists of a hollow-core photonic crystal fiber (PCF), large core fiber (LCF), a resonant fiber-scanner (RFS), a gradient-index lens system (GRIN), micro-beamsplitter prisms (BSP), photomultiplier tube, laser source and sample (S).

An ultrafast Ti-Sapphire pulse laser (812 nm wavelength, 100 fs pulse width and average power of about 300 mW, shown in red in Figure 1.5(a)) was used for excitation. The laser pulses were coupled to the PCF and guided to the fiberscope headpiece. RFS was controlled by external piezo drive signals (PDS) to scan the fiber cantilever spirally. The BSP was employed to conduct the laser into the GRIN lens, which focus the laser onto sample. The fluorescence emission (green) passed through the GRIN lens, BSP and then was guided by the LCF to photomultiplier tube. Two-photon image of 500 nm diameter fluorescent microspheres has been successfully obtained by the fiberscopy as shown in Figure 1.5(b). The lateral intensity profile of an example microsphere is shown in Figure 1.5(c). The profile shows a spatial resolution of about 1 µm. This fiberscopy was also successfully used to measure in vivo the calcium in rat brain. The fiber-based system is ultra-compact, sensitive, light (the weight is about 0.6 gram) and easy to handle. It is particularly suited for optical recording of neural network dynamics in awake.

Optical fibers have also been fitted onto SNOM systems for near-field fluorescence excitation and collection. Several kinds of fiber probes have been developed for high resolution SNOM imaging: aperture and active fiber probes in emission mode (as light nanosources) and double tapered aperture fiber probe in reflection mode (emission/collection through the same fiber tip). Fluorescence imaging down to single photon sources were success-
fully achieved with aperture probes [37] where active probes behaves as single fluores- cent sources to locally illuminate (non fluorescent) sample surface. In the latter case, diffracted fluorescent signal is collected through an objective.

Several active fiber tips with emitters attached at the apex have been reported for scan- ning near field optical microscopy (SNOM) imaging. Fluorescent terrylene molecules [38], NV diamond [39], QDs [40] have been grafted at the very tip. In 2008, Huant’s group proposed a fluorescent oxide nanoparticles integrated active fiber tip for near field imaging[41]. In this report, rare-earth doped oxide $Y_{2}O_{3}$ : $Ce^{3+}$ nanoparticles with the diameter of about 10 nm and stable fluorescence were integrated to the apex of chemi- cally etched single mode fiber tips. The oxide nanoparticles were excited with 458 nm line $Ar – Kr$ continuous mode fiber laser directly from the fiber tips, the fiber was thus used to excite the fluorescent source. Emission occurred at $\lambda = 560$ nm. The fiber tip was deposited with an aluminum layer and an aperture of 400 nm diameter was etched at tip apex. Then luminescent nanoparticles were sprayed directly into the aperture as shown in Figure 1.6(a). This probe has been demonstrated in the nano-imaging of 400 nm wide gold discs (Figure 1.6(b)).

Figure 1.6: (a) Electron micrograph of a metal-coated optical tip subsequently loaded with YAG cluster deposited directly. (b) Near-field optical image acquired simultaneously on a sample consisting in gold discs (400 nm diameter deposited on a silica cover slip.) [41].

A special fiber has been proposed as SNOM probe for near field imaging [42]. The experiment setup is shown in Figure 1.7(a). A fiber tip with double-tapered structure was first fabricated by chemical (hydrofluoric acid solution) etching. After 140 nm thickness Au metal film was deposited onto the fiber tip. A small aperture was formed by pouncing the probe against a quartz substrate and squeezing the Au out to the side. Using this method, well defined small aperture with a diameter of 20 – 50 nm can be created. The fiber probe was used both for illumination and fluorescence collection in this experiment. A sample was prepared by spin coating single dye molecules (excited by 633 nm He-Ne laser and emit fluorescence with wavelength of 700 nm) onto a quartz substrate. The fiber-sample distance was controlled by a shear-force feedback system. Using this structure, near-field images of single molecules has been achieved for the first time with both emission and collection through the same fiber tip (Figure 1.7(b)). A high resolution of 15 nm, which is smaller than the probe aperture size, has been obtained which is due to the nonradiative energy transfer between molecules and probe.
Besides the application for SNOM imaging, efficient collection of the fluorescence signal from single photon emitters is a research hotspot with potential applications in quantum information processing and telecommunications. Several processed fibers have been proposed to realize direct in-fiber collection of single photon emission with high efficiency.

A theoretical concept of tapered fiber was first proposed by Davanco’s group to be applied in single photon collection in 2009 [43]. Later on, experimental studies based on this idea have been reported [44, 45, 46]. In 2011, the first experimental demonstration of single emitter fluorescence collection was reported by Fujiwara’s group [47, 48]. In their report, a fiber with 300 nm diameter taper was fabricated by heat pulling. The tapered fiber was used to detect the photons from CdSe/ZnS QDs (9.8 nm diameter) whose emission wavelength is at 620 nm and excitation wavelength is at 543.5 nm. An 0.8-NA lens was employed to focus the laser to excite the QDs as shown in Figure 1.8(a) and (b). The experiment demonstrated a total fluorescence coupling efficiency of 7.4%. In 2009, Davanco’s group proposed a new theoretical model of tapered fiber with a waveguide integrated [49]. A single emitter was embedded inside the waveguide as shown in Figure 1.8(c). The excitation laser was input from one fiber end and the fluorescence can be detected at both the two fiber ends. A detection efficiency of 70% was predicted. This scheme needs to be experimentally demonstrated.

The demonstrated experiments about the tapered fiber single photon detection are based on objective excitation. But there are also fiber systems developed for simultaneous single emitter excitation and single photon detection.

In 2011, a micrometer-scale single photon source was experimentally demonstrated by Schoder [50]. It consists of a single NV-center diamond emitter on a photonic fiber as shown in Figure 1.9. In the experiment, a commercial photonic crystal fiber with outside cladding diameter of 90µm and core diameter of 1.5µm was employed. A 30 nm nanodiamond was deposited at the core of the photonic crystal by pick-and-place manipulation at room temperature (AFM technique). The nanodiamond was excited at a wavelength of 532 nm. The excitation and collection were realized by the same fiber. The experimental collection efficiency was shown to be equivalent to that of a 0.82-NA objective. This alignment-free fiber-based single photon source can be easily integrated into fiber networks for quantum cryptography or quantum metrology applications.
1.2. **FIBER SYSTEM FOR LUMINESCENCE DETECTION**

![Image](image.png)

Figure 1.8: Tapered fiber as a collection tool of NE fluorescence. (a) The scheme of the tapered fiber in collection mode. (b) The experiment scheme [47]. (c) The scheme of hybrid waveguide which combines a taper fiber and a waveguide [49].

![Image](image.png)

Figure 1.9: The photonic crystal fiber used for excitation/collection of fluorescence from single nanoemitter [51].

An open cavity that consists of a fiber-bonded micromirror of moderate reflectivity and a planar distributed Bragg reflector with GaAs QDs embedded in between has been proposed [51]. The scheme is shown in Figure 1.10. For the fiber-bonded micromirror, the multilayer coating undergo a slight dip right at the fiber core. The coating layers consisted of a release layer and SiO$_2$/TiO$_2$ dielectric stack. The planar Bragg reflector included 32 pairs of alternating layers of GaAs and AlAs and a GaAs spacer. The QDs were distributed at the center of the GaAs layer. In the experiment, He-Ne laser or picosecond pulsed Ti:sapphire laser at 790 nm was used to excite the QDs. The emission wavelength range of the QDs was 880 – 980 nm. The sample was connected to a piezo-electric actuator. The distance between the micromirror and Bragg reflector was precisely tuned by a voltage applied to the piezoactuator, so the cavity can be modulated to be mode matched to the single mode fiber. With a cavity length of about 10 µm, 10% photons from a single...
QD have been collected into the fiber at 7K temperature environment.

Figure 1.10: The schematic of external-mirror microcavity [51].

High refractive index nanowires have been proposed as nanostructure to ensure tight lateral confinement of guided modes [52, 53] as well as the fluorescence photons from embedded quantum dots. They have been investigated to extract and guide single photons for far-field emission into reduced solid angle ranges (enhanced emission directionality) more compatible with efficient fiber coupling, and thus fibered single photon sources [54].

An optical fiber integrated photonic wire has been proposed for efficient single photon detection through an optical fiber. The photonic wire was made of GaAs with a refractive index of 3.45 as shown in (Figure 1.11(a)) [54]. A self-assembled InAs QD was embedded 110 nm away from the nanowire sharp end. The wire was glued onto the core of a standard fiber as shown in Figure 1.11(b) (fiber core diameter of 4.4µm, fiber core refractive index of 1.4563 and fiber cladding refractive index of 1.4513). In the experiment as shown in Figure 1.11(c), a continuous laser diode source at 830 nm was employed to excite the QDs and the emission wavelength was at around 969nm. The experiment was performed at cryogenic temperature. A collection efficiency at the output of the fiber of about 5.8% has been realized.

Finally, optical fibers have been used to experimentally detect the local X-ray excited optical luminescence of a sample [55]. In the experiment shown in Figure 1.12(a), a SNOM system was developed to achieve chemical mapping at nanometer-scale resolution sample thanks to their X-rab in-fiber luminescence collection from the sample. The fiber tip was coated by aluminum layer and a 70 nm diameter aperture was opened at the apex. The fiber probe was aligned with the X-ray beam center and the sample scanning was realized by moving the sample. By recording the images with X-ray energy below and above Zn – Ka absorption threshold at 9600eV (Figure (f)) and 9664eV (Figure (g)), respectively, the areas where element Zn locates are emphasized. Correspondingly, by recording the images with X-ray energy below and above W – L3 absorption threshold at 10190eV (Figure (h)) and 10207eV (Figure (i)), respectively, the element W region is imaged. Figures (b)-(e) are the topography images of the sample, which indicates that same sample area was scanned during the four scanning.

The comparison of the properties between optical lens systems and optical fiber systems for luminescence detection can be expressed by the table 1.1:
1.3. NANOANTENNA-ASSISTED SUPER EMITTER

Nanoantennas can be placed in local environment of single photon emitters to tailor single photon emission [57]. The fluorescence can be enhanced and the emission directionality can be tailored due to the influence of nanoantennas. The union of an emitter and a
nanoantenna can be considered as a “super emitter” with much strong fluorescence or high emission directionality, thus the detected fluorescence signal from this kind of emitters will be improved. Most of super emitter addressing/collection scheme is realized in far-field with confocal optical microscopy architecture.

It has been demonstrated that gold nanoparticles support highly localized plasmon resonances when excited by external illumination. They have been used as nano-optical antennas with the ability of both enhancing excitation and emission rates of nearby fluorescent emitters [26, 58, 59]. In general, plasmon oscillations of metal particles can be developed in terms of multipole moments, whereby the dipole component is the dominant contributor if the particle diameter is much smaller than the radiation wavelength [60]. Metal nanoparticles thus behave as dipole antennas [61] but with induced (and not fixed) dipole moment oriented by the excitation field, due to spherical symmetrical of the particles. By enhancing both excitation and fluorescence emission rates, such antennas improve the far-field fluorescence detection usually realized with spherical optics. Theoretical analysis and different kinds of experiments have been developed to demonstrate the influence of gold nanoparticle on emitters’ luminescence [26, 58, 59, 62].
1.3. NANOANTENNA-ASSISTED SUPEREMITTER

Figure 1.13: Two experiments of metal nanoparticle antenna. (a) a single gold nanoparticle is scanned across a single molecule. The green lines sketch the inhomogeneous excitation field. (b) The schematic of the experimental arrangement. (a) and (b) are cited from [59]. (c) Experimental setup for simultaneous far-field optical detection and near-field manipulation of individual diamond nanocrystals and gold nanoparticles. (d) AFM images of different gold-diamond configurations. (c) and (d) are cited from [58].

A gold nanoparticle has been employed by Kuhn’s group to reach a fluorescence detection enhancement of more than 20 times [59]. At the same time, they found a 20-fold shortening of the excited state lifetime. In the experiment, Terylene molecules with excitation wavelength of 532 nm and emission wavelength of 600 nm were spin coated into thin para-terphenyl (PT) crystalline film with a thickness of 20 – 30 nm. Gold particle with a diameter of 100 nm (Figure 1.13(a)) was fixed onto a heat-pulling fiber tip and controlled by a shear-force stage (Figure 1.13(b)). A confocal objective with numerical aperture of 1.4 was used to excite the molecules and collect the fluorescence. A similar experiment has been performed with two gold particles placed in proximity of an emitter. A fluorescence detection enhancement of 18-fold has been achieved [58].

A plasmonic antenna, which was composed of a single nano aperture at the center surrounded by several concentric grooves, was proposed to control the emission directionality of molecules in water solution [63]. The experimental scheme is shown in Figure 1.14(a). In the experiment, nano molecules (Alexa Fluor 647, Invitrogen, Carlsbad, CA), which were excited by 633 nm laser and emit photons at 680 nm, were dispersed inside liquid. The plasmonic antenna was based on a glass coverslip with 190 nm thick Au layer and 60 nm thick chromium layer deposited on it. A central aperture with 140 nm diameter, grooves with a width of 200 nm and depth of 65 nm have been etched to form the grating. 5 circular grooves have been formed with a period of 440 nm. The liquid sample was distributed at the grating center. An objective with a numerical aperture (NA) of 1.2 was used to excite the sample and detect molecule fluorescence.

The most important parameter of the antenna is the distance \( d \) between aperture center and the first groove, which determined the radiation direction. SEM images of the grating antenna with \( a = \lambda_{np} \) and \( a = \lambda_{np}/2 \) are shown in Figure 1.14(b), where \( \lambda_{np} \) is the
surface wave wavelength at the metal-dielectric corrugated interface. As a comparison, a single aperture plate without any grooves was fabricated. The experiment showed no directive emission using this plate as shown in Figure 1.14(c). When a equals to \( \lambda_{\text{dp}} \) (Figure 1.14(d)), a very directive radiation with an extension angle of \( \pm 14^\circ \) was realized. But for the \( a \) taken as \( \lambda_{\text{dp}}/2 \), the radiation points toward \( \pm 30^\circ \) with a minimum intensity in the direction normal to the sample (Figure 1.14(e)). The difference is due to the interferences between the emission directly from the central aperture into far-field and the surface-wave coupled emission which was radiated by the grooves toward far-field. Besides the highly directional fluorescence emission, the concentric rings structure can also further focus the light field at aperture center, thus the excitation rate would be enhanced. The modulated emission directivity and the enhanced excitation/emission rate lead to a fluorescence detection enhancement with a factor up to 120 fold [63].

Yagi-Uda antenna was first applied in radio wave detection or emission and a conventional Yagi-Uda antenna in radio wave regime is shown in Figure 1.15(a) [64]. It was then applied to nano-optics by Kosako's group [64] and Van Hulst’s group [65]. In nano optics, Yagi-Uda antenna is generally constituted by five carefully designed metal rods in a row, with one larger-sized rod located at one end of the row as reflector, one rod close to the "reflector" rod taken as the feed where emitters locate, and three smaller-sized rods in another side of the row playing a role of directors. They can emit photons from dipolar fluorescent sources with highly improved directionality. Van Hulst’s group fabricated golden rods on a glass substrate as Yagi-Uda antenna [66]. SEM image of the antenna is shown in Figure 1.15(b). The QDs were excited by 633 nm laser with emission wavelength larger than 830 nm. 1.46-NA objective was employed to image the QDs. The confocal images of the Yagi-Uda antenna-based QDs are shown in Figure 1.15(c) and the radiation pattern of an example QD is shown in Figure 1.15(d). The radiation pattern clearly shows directional QD emission. In addition, Yagi-Uda nano antenna can not only guide the emission into one direction, it can also enhance the excitation and emission rates [65, 67].
1.3. NANOANTENNA-ASSISTED SUPEREMITTER

![Diagram of Yagi-Uda antenna](image)

Figure 1.15: The application of Yagi-Uda antenna into nano optics field. (a) the principle of Yagi-Uda antenna in radio wave regime [64]. (b) SNOM image of a Yagi-Uda antenna in nano optics. (c) Scanning confocal luminescence QDs images of Yagi-Uda antenna. (d) Radiation pattern of one example QD in (c). (b), (c) and (d) are cited from [66].

Bowtie nano-antennas are composed of two face-to-face metal triangles which are separated by a nanometer scale gap as shown in Figure 1.16(a). The antenna is polarization selective, its typical anisotropic elongated shape originates an oriented electric dipole moment. It is resonant with the linearly polarized light whose polarization is parallel with the two BNA triangles. Bowtie antenna can generate a nanoscale confined and enhanced light spot when excited optically at resonant wavelength with the right incident polarization. The confined light spot would disappear when the excitation light polarization is rotated by 90°.

![Diagram of Bowtie antenna](image)

Figure 1.16: The application of bowtie antenna in fluorescence enhancement. (a) Bowtie antenna scheme coated with TPQDI molecules in PMMA on a transparent substrate. (b) SEM image of a Bowtie antenna. (a) and (b) come from [68]. (c) SEM image of end surface a bowtie antenna fabricated at the end of AFM tip. (d) SEM image of the side surface of Bowtie antenna. (c) and (d) come from [69].
Bowtie nano-antenna has been experimentally adopted in single emitter fluorescence enhancement. Moerner’s group fabricated an array of gold bowtie antennas coated with TPQDI molecules (excitation/emission wavelength: 780nm/850nm) in PMMA and the sample was scanned by confocal microscopy [68]. The sample with bowtie antenna was shown in Figure 1.16(a) and the SEM image of the bowtie antenna was shown in Figure 1.16(b). The experiment demonstrated a fluorescence enhancement up to a factor of 1340 due to greatly enhanced absorption and increased radiative emission rate [68]. Hecht’s group fabricated a 40nm thickness aluminum bowtie antenna onto an AFM tip and the antenna was put into emitters’ local environment to enhance the fluorescence emission (Figure 1.16(c) and (d)). Emitters with 532nm excitation wavelength and 580nm emission wavelength were employed. The distance between Bowtie antenna and emitters was controlled by AFM system. The result demonstrated clearly reduced excited-state lifetime (enhanced emission rate) [69].

Similarly to Bowtie nano-antenna, Bowtie nanoaperture antenna (BNA) is composed of two face-to-face triangle apertures etched inside metal film which are separated by a nanoscale gap. BNA holds the optical properties of the bowtie nano-antenna, in terms of light spot confinement and enhancement and polarization sensitivity. Gong’s group applied the aperture antenna to enhance single photon detection and studied the influence of BNA to excited-state lifetime of emitters [70]. In his experiment, BNA was fabricated by focused ion milling (FIB) into 170 nm aluminum films as shown in Figure 1.17(a). Then fluorescent molecules (excitation/emission wavelength: 633 nm/670 nm) were coated onto the thin metal film. Optical objective was used to collect the fluorescence as shown in Figure 1.17(b). The experiment result showed a fluorescence enhancement of 7 fold and the lifetime of the excited state changes obviously with two perpendicular polarization light excitation.

![Figure 1.17](image-url)
1.3. NANOANTENNA-ASSISTED SUPEREMITTER

antenna. First, it is a background free when used in transmission mode, i.e. the detected signal comes exclusively from the nano-antenna resonance because the opaque surrounding metal layer stops the incident field that does not interact with the nanoantenna resonance. Second, it can be more easily fabricated at the end of a metal coated tip as less metal needs to be removed to form the nano-structure as compared to the bowtie nano-antenna. BNA have thus been implemented at the end of cantilever SNOM probes for near-field imaging [71], photolithography [68]. They have also been proposed as a key-connection (two-way communication channel) between "nanoscale world" and an optical fiber [72], with direct application in fiber SNOM imaging (collection mode) [71, 72], fiber nano-optical tweezers [73, 74] and sensing [75].

The concept of triangle aperture was proposed in 2002 showing a strong field enhancement at only one rim when illuminated with light of proper polarization [76]. In 2005, the aperture was integrated to a SNOM probe to be placed in proximity with fluorescent molecules for high resolution imaging [77]. The aperture fabrication was based on a tetrahedral glass body. The common glass tip was coated by aluminum layer and then a 50 nm-sized triangle aperture would be formed onto the tip apex by FIB. The SEM image of a fabricated triangle aperture is shown in Figure 1.18(a). A focused laser beam at 633 nm illuminated the aperture through the glass body and a highly enhanced and confined light spot would be generated. Terrylene dimide(TDI) molecules embedded in a 10 nm thick PMMA layer were prepared onto a clean cover glass as the sample. The triangle aperture was placed in near field region of the sample and the transmitted light from the aperture was used to excite the molecules. The molecule fluorescence was collected by a 1.3 NA oil-immersion objective and finally detected by an avalanche photon diode. The near-field fluorescence image of randomly oriented TDI molecules is shown in Figure 1.18(b) and the spatial imaging resolution of 30 nm has been realized.

![Figure 1.18: (a) SEM image of a triangle aperture. (b) Scanning images of fluorescent molecules with triangle aperture excitation [77].](image)

The nanoantennas, such as monopole antenna [78] and BNA [79], have been integrated with optical fibers to offer enhanced nanosource for emitter excitation. In 2014, a hybrid antenna of the combination of BNA and monopole antenna was proposed by Garcia-Parajo’s group to realize dual-color imaging with high resolution [80]. The hybrid antenna
was fabricated onto a heat-pulling fiber tip apex (SM600, Fibercore) which was coated by 130 nm aluminum layer. A BNA was carved into the metal layer at the fiber apex and a monopole antenna with a length of 60 – 80 nm was fabricated beside an edge of the BNA gap as shown in Figure 1.19(a). A SEM image of the hybrid antenna is shown in Figure 1.19(b). To realize the dual color nanoimaging of individual molecules with high resolution, the mixture of DiD and DiI fluorescent molecules sample was prepared. The two kinds molecules with concentration of $10^{-6} M$ were mixed with PMMA and the mixture was spin coated onto glass substrate. The corresponding excitation wavelengths of DiD and DiI molecules are 560 nm and 633 nm, respectively. A Laser source including the two excitation wavelengths components was input from the fiber. A very confined and enhanced light spot would be formed at the end of the hybrid antenna as a nanosource to excite the two emitters. An oil-immersion optical objective ($\times 100$, 1.3 NA) was used to detect the luminescence in far field as shown in Figure 1.19(c). Two APDs were employed to detect different wavelength fluorescence from the two emitters. The multi color scanning images of the two emitters with a high resolution of 20 nm have been obtained as shown in Figure 1.19(d)). In this imaging process, monopole antenna with diameter of 25 nm and length of 80 nm was employed.

![Figure 1.19](image)

Figure 1.19: (a) Scheme of the hybrid antenna. (b) SEM image of the hybrid antenna. Scale bar: 300 nm. (c) Experimental setup scheme. (d) Near-field images of the sample. Scale bar: 200 nm [80]

The applications of nanoantennas in X-ray excited luminescence has yet been reported.

1.4/ CONCLUSION

In this chapter, I introduced the luminescence detection techniques. Optical lens-based luminescence detection, optical fiber-based luminescence detection and nanoantenna-assisted super emitters have been included in this chapter. The optical lens system, especially microscopy was the first technique in luminescence detection with the advantages of high resolution, improved detection efficiency and so on. The classical examples of lens systems in luminescence detection have been reviewed in the first section. But application of the lens systems are still limited due to the bulky optical systems, strict
alignment requirement and expensive optical elements. Fiber system is an alternative with the advantages of flexible, compact, easy-to-control and low-cost. The fiber applications in luminescence detection have been included in the second section. At the end, nanoantenna-assisted super emitters was described at the third section. The super emitters have been applied with microscopy systems for enhanced single photon detection. But due to the limitations of the bulky lens systems, it is promising to combine the super emitters and fiber system from photon detection, which is one of the most important parts in our job.
This chapter is focused on the direct far-field coupling of fluorescent emitters to a single mode optical fiber. We intend here to generate far-field fluorescence imaging bench that is ultra-compact (i.e. free from bulky optics), plug-and-play, and showing new properties. We thus propose to realize this optical interconnection between the fluorescent samples and the fiber with Bessel beams which are known to be non-diffracting beams [81]. Bessel beams have the unique ability of confining light up to the diffraction limit over distance several orders of magnitude larger than the Rayleigh distance of gaussian beams [29]. The non-diffracting nature of Bessel beam has opened new perspectives in a large panel of scientific and industrial domains such as optical acceleration [82, 83], particle guiding [84] and manipulation [85, 86], nonlinear optics [87, 88, 89, 90, 91, 92, 93], optical interconnection and alignment [94, 95], imaging [96, 97, 98, 99], microfabrication [100, 101] and lithography [102]. The use of Bessel beams allows for dramatically extending depth-of-field of imaging systems [21]. The engineering of fibered high depth-of-field fluorescent imaging could be of interest in scientific, medical and industrial domains. Here, we intend to generate all-fiber imaging process, including both fluorescence excitation and collection by exploiting reciprocity principle. The resulting imaging bench would therefore be ultra-compact, flexible, and plug-and-play, thus usable out of the lab by non specialists in optics, and compatible with endoscopy.

The direct generation of Bessel beams from a fiber mode remains a real challenge. Usually, Bessel beams are produced with bulky optics such as holograms, diffractive elements, spatial light modulator, axicons and lenses, etc [103, 104]. Axicons have however been already successfully reported at the end facet of an optical fiber, by different fabrication techniques such as fiber chemical etching, polishing, FIB (Focused Ion Beam) milling, resulting in the direct generation of Bessel beams at the end of single mode optical fibers [105, 106]. However, the Bessel beam aspect ratio (width versus length of the central light confinement) stays modest in these cases. This is explained by the fact that the length of the Bessel beam is limited by its overall width (geometrical optics consideration, see Ref.[107]), and in the above cited configuration the aspect ratio of the Bessel beam is limited to a few tens of microns by the micron size lateral extension of the fiber mode. One way to enhance Bessel beam length consists of enlarging the impinging wave onto the axicon, leaving the idea of a direct axicon engineering at the end facet of a fiber. Since the fiber output can be seen as a point-like source, inserting a lens in between an axicon and a fiber so that the fiber end is positioned at focus of the lens seems to be a very simple solution to turn the fiber emission into a Bessel beam of very high aspect.
ratio (see Figure 2.1). In other words, the generation of a Bessel beam from a single mode fiber can be realized with a simple optical bench that consists of an axicon and a collimating lens. The challenge is however to achieve an ultra-compact bench which is auto-aligned with respect to the fiber, thus to avoid bulky optics.

![Figure 2.1: Scheme of the generation of a Bessel beam from a single mode optical fiber.](image)

In this chapter, we propose a solution to this issue with the development and study of the properties of AXIGRIN, a new tool which combines the diffraction properties of an axicon and a collimator in a single element in direct contact to a fiber. It is achieved by engineering one of the two end facets of a GRadient INdex (GRIN) lens in a conical (Axicon) shape. The resulting integrated optical bench is compact (of the millimeter range), auto-aligned with respect to the fiber and plug-and-play. In the first section, we will show the concept and the underlying theoretical background of the AXIGRIN by studying axicon properties and GRIN lens properties. The fabrication of the new tool will be detailed in the same section. Bessel beam generation (zeroth-order and first-order Bessel beam) will be demonstrated in the second section. Experimental process and Bessel beam properties will be analyzed. Then, fiber AXIGRIN will be demonstrated in fiber fluorescence imaging using infrared colloidal QDs (PbS). The general purpose of this chapter is shown in Figure 2.2.

### 2.1/ Theory and fabrication of an AXIGRIN

In this section, we propose the new tool, i.e. an axicon fabricated on one end facet of a gradient index lens (GRIN lens), to generate a Bessel beam from a fiber mode. The resulting optical system is compact (no bulky optics is used), simple and flexible, which is of practical interest for end-users. This section is divided into two parts: basic theory of AXIGRIN and fabrication process. In the first part, the theoretical basis of Bessel beam generation with the principle of axicons and GRIN lens is explained. The second part is about the fabrication process of the AXIGRIN.
2.1. THEORY AND FABRICATION OF AN AXIGRIN

2.1.1/ BASIC CONCEPT/THEORY OF THE AXIGRIN

Axicon is a typical and mature tool to generate Bessel beam from a collimated optical beam and GRIN lens is used to generate collimated optical beams from a point-like light source. By combining these two components in a single one, we propose an AXIGRIN to generate a Bessel beam from the output of a single mode fiber.

BASIC THEORY OF AXICONS AND BESSEL BEAMS

Zemanek's group gave detailed theoretical analysis of the Bessel beam generation from a Gaussian beam by usual (linear) axicons in [108, 109]. I will expose here basic theory about paraxial Bessel beams and axicons. Figure 2.3 shows the principle of Bessel beam generation from a collimated Gaussian beam by an axicon. Ideally, an axicon is a conical lens, or axis symmetrical prism, which generates a distribution of plane waves whose wave vectors lie onto a fictive cone. All these plane waves couple and give rise to an interferogram whose field distribution is described by Bessel functions (axis symmetrical cosines). The electric field distribution of ideal Bessel beams (infinitely extended, non physical) in the paraxial regime are described by:

\[ E(r, z) \propto J_0(\alpha r) \exp(\text{i} wz), \]  

(2.1)

for the zero order Bessel beam and,

\[ E(r, z) \propto J_1(\alpha r) \exp(\text{i} wz) \]  

(2.2)

for the first order Bessel beam. Zero-order Bessel beam is expressed with scalar diffraction theory [29]. Here, \( \alpha \) and \( w \) are constants described by \( \alpha^2 + w^2 = (2\pi/\lambda)^2 \) where \( \lambda \) is the wavelength.

In the case of real Bessel beams of limited extensions, the above described field distributions can be apodized with square integrable functions. The more realistic apodization function is the gaussian function which leads to the well-known Bessel-Gauss beams.
generated from Gaussian beams [110]. Depth Of Focus (DOF) of an axicon defines the longitudinal length of the real Bessel-Gauss beam within which the on-axis intensity keeps unchanged theoretically. Generally, DOF is decided by the radius of incident (Gaussian) beam $\omega$, the axicon angle $\alpha$ and the refractive index $n$ of axicon material as shown in Figure 2.3. $\alpha_0$ is the refractive angle of the incident beam and $\theta$ is the angle between the edge refracted light and the optical axis. For the axicon with large apex angle and very small $\alpha$, $\alpha_0 = n\alpha$ according to Snell’s law [111] and thus, $\theta = (n - 1)\alpha$. According to the simple geometrical description of the Bessel-Gauss beam generation, DOF can be depicted as the function below:

$$DOF = \frac{\omega}{\tan\theta}$$  \hspace{1cm} (2.3)

According to the relationship between $\theta$ and $\alpha$, $n$, we can get that:

$$DOF = \frac{\omega}{\tan(n - 1)\alpha}$$  \hspace{1cm} (2.4)

To make this description in accordance with the optical wave description, the DOF can be redefined as the length of the interval, where the on-axis intensity is larger than $0.3227$ fold of the maximal value. The maximal value of the on-axis field intensity occurs at the longitudinal position:

$$z = \frac{\omega}{2 \tan(n - 1)\alpha}$$  \hspace{1cm} (2.5)

The largest width of the Bessel beam is noted as $h$ in Figure 2.3:

$$h = \omega$$  \hspace{1cm} (2.6)

For an ideal Bessel beam, the on-axis intensity remains constant with propagation. But for experimentally generated real Bessel beam, the finite aperture of axicons results in
an on-axis intensity that varies with propagation distance and ultimately, a limit to the diffraction free range [112]. The on-axis intensity of the Bessel beam change along z direction is depicted in the Figure 2.4:

![Figure 2.4: On-axis intensity for \( \alpha = 2.5^\circ \), \( \omega = 2.5 \) mm (width of the incident gaussian beam) and \( \lambda = 532 \) nm [109].](image)

The generation of Bessel beams with axicons is however often limited to bulky optics mounted into optical benches that are difficult to align for end users who are not specialists in optics. It thus remains a key challenge to generate Bessel beams in compact, versatile and flexible devices. The production of micro-axicons allowed for unprecedented system miniaturization meanwhile avoiding preliminary optical alignment [105, 107, 113, 114, 115]. However, axicon size reduction down to the microscale is irremediably accompanied by a strong limitation of the Bessel beam length that do not exceed a few tens of micrometers (due to the limited lateral extension of the axicon of a few microns).

**Basic theory of GRIN lenses**

The GRIN lenses are glass cylinders described with a radially varying refractive index, as expressed in 2.7:

\[
\begin{align*}
    n &= n_0 \left[ 1 - (k/2)r^2 \right] \\
\end{align*}
\]  

(2.7)

where \( n_0 \) is the index at the center of the lens, \( k \) is a parameter called Gradient Constant, and \( r \) is the radial coordinate [116]. The refractive index is maximum at the GRIN center and decreases as \( r \) increases. Such an index distribution leads to oscillating sinusoidal light rays within the lens, as shown in Figure 2.5 for one full sinusoidal path (one "pitch"), 0.25 pitch GRIN lens and 0.23 pitch lens.

We can see from the Figure 2.5(a) that a light spot at the entrance facet of the one-pitch long GRIN lens can be reconstructed at the rear facet of the lens. The gradient refractive index inside the lens originates the sinusoidal optical path. If we use 0.25-pitch GRIN lens as shown in Figure 2.5(b), an incident collimated beam is focused onto the rear facet.
CHAPTER 2. FLUORESCENCE IMAGING WITH A FIBER AXIGRIN

Figure 2.5: (a) light optical path inside a GRIN lens with a length of one full sinusoidal path (one pitch). (b) light optical path inside a 0.25 pitch GRIN lens. (c) light optical path inside a 0.23 pitch GRIN lens.

of the GRIN lens. The image focal plane of the 0.23-pitch GRIN lens (Figure 2.5(c)) is positioned after the rear facet of the GRIN lens, which avoids high power confinement within the GRIN lens. According to reversible property of optical lens, the 0.25-pitch and 0.23-pitch lenses can also be used for collimating the waves emitted by a point-like source (for instance, the spot at end of an optical fiber) placed at, or close to, the entrance facet of the GRIN lens.

The pitch length of a GRIN lens is expressed as:

\[ p = \frac{2\pi}{\sqrt{k}} \]  

(2.8)

According to the function 2.8, we can see that the GRIN lens pitch is defined by the Gradient Constant \( k \), which describes the refractive index variations of the GRIN lens.

THE DESIGN OF AXIGRIN

The AXIGRIN principle is to generate a Bessel beam from a point-like source (the end of an optical fiber) by transforming one of the two flat end facets of a 0.25-pitch GRIN lens into a cone, i.e. an axicon (see Figure 2.6(a)).

The operation principle of the 0.25-pitch and 0.23-pitch AXIGRIN is depicted in 2.6(b) and (c), respectively. The first configuration (Figure 2.6(b)) allows for easy positioning of a point-like source (in our case, the fiber end facet) in contact to the GRIN lens whereas the second one (Figure 2.6(c)) protects the GRIN lens from possible damages produced by high power point-like sources. In both cases, a collimated beam is projected onto the output conical interface of the AXIGRIN, leading to a transmitted Bessel beam.

2.1.2/ FABRICATION OF THE AXIGRIN

To obtain the above described AXIGRIN, an axicon is fabricated on a GRIN lens with a conventional mechanical polishing procedure. First, a rough cone is ground at the output facet of the lens with a spinning abrasive wheel mounted onto a rotation-translation stage (Figure 2.7(a)). The cone angle \( \theta \) of the axicon is simply defined by the tilt angle \( \theta/2 \) of the fine abrasive wheel with respect to the GRIN axis. We chose a cone angle of 82°, leading to a numerical aperture (NA) of the AXIGRIN of about 0.085. Then, the cone is polished manually with a series of 5 diamond pastes (from Toutoy & Bertholon) of decreasing diamond grain sizes (grade 30 down to grade 1) applied successively onto the
To investigate the properties of the AXIGRIN, we design two different experiments. The first experiment, conducted at 633 nm, is aimed at demonstrating zeroth-order and first-order Bessel beam generation with direct imaging technique with the 0.25-pitch AXIGRIN. In the second experiment, we investigate the fluorescence imaging property of the AXIGRIN (0.23-pitch at 1550 nm) using infrared PbS QDs whose emission wavelength is centered at 1500 nm. This technique is compared with direct imaging with a cleaved optical fiber. The AXIGRIN is demonstrated and characterized both in collection and excitation/collection modes.
2.2.1/ BESSEL BEAM DEMONSTRATION

We demonstrate the Bessel beam generation with an AXIGRIN (0.25 pitch GRIN lens at 633 nm) connected to an optical fiber coupled to a HeNe laser (λ = 633 nm) [?]. The emission area at the fiber end facet is limited by the fiber core diameter to a value smaller than 30µm² and can therefore be considered as a point-like source for the AXIGRIN. This point-like source is necessary for the AXIGRIN to generate a Bessel beam. The fiber end facet is self-aligned and positioned with respect to the AXIGRIN by means of a simple commercial pigtailed ferrule device (Thorlabs, model SMPF0206). Figure 2.7(f) shows a picture of a resulting fiber AXIGRIN. The AXIGRIN dimensions are that of the GRIN lens: 1.8 mm wide and about 4.6 mm long. The overall pigtailed AXIGRIN is 2.8 mm large and 10 mm long. Smaller fiber axicon bench can be however achieved by directly gluing the fiber to the AXIGRIN. Note that smaller GRIN lenses are also commercially available with diameter of 1 mm or smaller (instead of 1.8 mm in our case). Therefore very compact pigtailed axicon benches can be reached with the concept of AXIGRIN.
2.2. EXPERIMENTAL STUDY OF THE AXIGRIN PROPERTIES

**ZERO-ORDER BESSEL BEAM**

To generate a linearly polarized zeroth-order Bessel beam, we use an optical fiber operating in single mode regime at wavelength around 630 nm. The experimental setup is shown in Figure 2.8. The single mode fiber at $\lambda = 630$ nm is coupled to a Babinet-Soleil polarizer (Newport, model F-POL-IL, not shown in the setup) to launch a linearly polarized mode onto the AXIGRIN. An objective ($40 \times, 0.65\text{NA}$) is coupled to a charge couple device (CCD) camera. The fiber AXIGRIN is attached parallel to the optical axis, called (0z) in the following, so that the axicon faces the objective. It is mounted onto a precision 3D translation stage allowing AXIGRIN alignment with respect to the imaging system and displacement along (0z).

![Figure 2.8: Scheme of the experimental set-up: optical characterization of a fiber AXIGRIN](image)

The imaging bench is first calibrated with white light imaging of a transmission calibration grating so that the Bessel beam optical dimensions can be measured directly from optical images. To this end, the grating with the dimension of 300 grooves per millimeter is imaged with the objective and the distance between the objective and CCD camera is fixed to ensure the magnification of the optical system unchanged. Two images are recorded with the grooves oriented vertically and horizontally. Figure 2.9 shows the image of the grating oriented vertically, we can see that 40 periods of lines are visible, indicating a 0.13 mm-large field of view (along the horizontal direction).

![Figure 2.9: Scheme of the experimental set-up: optical characterization of a fiber AXIGRIN](image)

Figure 2.10 shows the zeroth-order Bessel beam generated with AXIGRIN system. Figure 2.10(a) displays an image of the beam in a transverse plane perpendicular to the
propagation axis ($0_z$). A single confinement is obtained at the beam center that is surrounded by a series of concentric light rings. We see from Figure 2.10(b) that the beam intensity profile along the transverse plane (solid red curve) is described by Bessel function $J_0$ (dashed blue curve), which is consistent with Equation 2.2.

![Figure 2.10](image)

Figure 2.10: Zeroth-order linearly polarized Bessel beam produced with the fiber AXI-GRIN: (a) beam transverse cross section, (b) beam profile along the radial coordinate (comparison with theoretical prediction (dashed curve))

As known, one important property of axicon is long field depth. To demonstrate the field depth of the AXIGRIN, the AXIGRIN is moved away from the objective step by step. At each step, a 2D transverse cross-section of the beam is grabbed by the CCD camera and stored. Then, the beam maximum is measured for each image. This “slice by slice” imaging process is stopped when the AXIGRIN is far enough from the objective so that the beam central spot vanishes. When the distance between the AXIGRIN and the object plane of the objective is 0, 0.5 mm, 1 mm, 1.5 mm, 2 mm and 3 mm, the Bessel beam cross-sectional images are recorded and shown in Figure 2.11(a)-(f), respectively. The experimental results show a quasi unchanged light confinement of 2.65 $\mu$m (FWHM measurement) at the beam center along the overall beam length estimated to a value of 3.5 mm, which implies a beam aspect ratio that exceeds 1300. This Bessel beam length is more than 62 times larger than the Rayleigh distance of a Gaussian beam with the same confinement ability at waist ($1/e^2$ width comparison). Theoretical predictions achieved with a simple ray-tracing through the AXIGRIN (based on the fiber NA and GRIN intrinsic properties) give a Bessel beam length of 2.9 mm. Moreover, the relative difference between the beam FWHM measured experimentally and the ideal Bessel beam predicted by diffraction theory (see Equation 2.2) does not exceed 1%. These results unambiguously prove that the AXIGRIN generates a Bessel-Gauss beam in paraxial regime whose propagation length is dramatically increased as compared to the fiber systems proposed so far [105]. Figure 2.11(g) is the Bessel beam central spot on-axis intensity change along the longitudinal propagation axis($0_z$). The on-axis intensity increases with the increase of the distance and reach the maximum at a distance of 1 mm. Then it decreases with the increase of the distance. The experiment result agrees with the theoretical prediction as shown in figure 2.4.
First-order Bessel beam is achieved by coupling the radially polarized $T_{M01}$ fiber mode to the AXIGRIN. This doughnut mode is generated within a 830 nm single mode fiber which behaves as a few-mode fiber at $\lambda = 633$ nm, the operation wavelength in our study. The fiber radial polarizer is shown in the Figure 2.12(b), with the same procedure as depicted in [104]. The linearly polarized He-Ne laser beam is expanded and collimated. In order to selectively excite the $T_{M01}$ mode within the fiber and cancel the fundamental fiber mode $HE_{11}$ simultaneously, a $\pi$-step phase plate is inserted in the path of the collimated beam.

When the linearly polarized collimated beam passes through the step element, half of the beam is $\pi$ phase retarded with respect to the other half, leading to polarization reversal over half the beam cross-section (see Figure 2.12(b)). Then, the phase modified collimated beam is injected into the fiber with an objective. The fiber can propagate only 4 modes called $HE_{21}^\prime$, $T_{E01}$, $HE_{21}^\prime$ and $T_{M01}$ (annular modes). The relative orientation of the incident linear polarization and the straight edge of the phase element limits the mode excitation to two modes. When the step-phase element straight line is parallel to the incident linear polarization direction, only the modes of $T_{E01}$ and $HE_{21}^\prime$ are excited within the fiber, leading the degenerated mode ($LP_{11}$). When the straight edge is perpendicular to the linear polarization, $T_{M01}, HE_{21}^\prime$ mode will coexist inside the fiber at the same time.
CHAPTER 2. FLUORESCENCE IMAGING WITH A FIBER AXIGRIN

Since the radially polarized beam corresponds to the fiber mode $T_{M01}$, we need to put the step-phase element region separate edge perpendicular with the linear polarization direction.

Then a Babinet-Soleil polarization controller is used to make both the torsion and a fine compression of the fiber for single mode selection in a very precise and simple way. It is an empirical operation, the Babinet-Soleil polarization controller is adjusted to make the fiber output beam to be a well-defined annular beam.

Figure 2.12: The generation of radially polarized beam. (a) influence of the step-phase element to the fiber guiding modes. (b) experimental setup of the generation of radially polarized beam.

The so-produced radially polarized annular beam is shown in Figure 2.13. Figure 2.13(a) represents the annular radially polarized beam. Figure 2.13(b) shows two perpendicular profiles of the annular beam which cross at its dark center. Figures 2.13(c)-(f) show the images of the beam cross-section through a linear polarizer (beam analyzer), whose axis directions are indicated with white arrows on each figure. The two grain structure in (c)-(f) is independent of the polarizer direction, which indicates that the radial polarization purity of the output beam is perfect and all other modes have been canceled.

To get the first-order Bessel beam, the radially polarized fiber mode excites the AXIGRIN, as the figure 2.8 shows. With the imaging bench, the first-order Bessel beam is characterized as well (Figure 2.14).

Figure 2.14(a) displays a cross-section of the beam diffracted by the AXIGRIN, which clearly shows a tiny doughnut at the beam center. We see from Figure 2.14(f) that the beam profile (solid red curve) is proportional to Bessel function $J_2^2$ (dashed blue curve). Figures 2.14(b-e) show images of the Bessel beam transmitted through a linear polarizer, for 4 different polarizer directions indicated by white arrows in the figure insets. The two light spot distribution within the image, which follows the polarizer axis, proves our first-order Bessel beam is radially polarized.

2.2.2/ INFRARED FLUORESCENCE IMAGING WITH A FIBER AXIGRIN

In fluorescence imaging experiment, the QDs from Evident company and made from PbS with nanoparticle diameter of 9 nm are used as fluorescent emitters. The QDs are initially
dispersed in liquid toluene with a concentration of 22.7 nmol/ml. The picture of the product is shown in 2.15(a). The left figure was taken by visible camera while the right one was taken by infrared camera. The QDs picture taken by visible camera is totally dark due to the absorption of visible light by QDs. The absorption curve of the QDs is shown in Figure 2.15(b), an absorption peak is shown at the wavelength of about 1450 nm. The light with wavelength shorter than 1000 nm can also be absorbed efficiently. The emission curve is shown in Figure 2.15(c). The emission peak is located at the wavelength of about 1500 nm. In the experiment, we set the excitation wavelength at 808 nm.

**Experimental setup and the preparation of the sample**

In this part, we propose and demonstrate the AXIGRIN for fluorescence imaging in the infrared spectral domain with a compact and plug-and-play system. Infrared fluorescence is achieved here with PbS QDs from Evidot company. Our AXIGRIN setup is aimed at coupling infrared QD fluorescence at $\lambda = 1500$ nm to a $SMF-28$ fiber. To characterize
CHAPTER 2. FLUORESCENCE IMAGING WITH A FIBER AXIGRIN

Figure 2.14: First-order radially polarized Bessel beam generated with a fiber AXIGRIN: (a) beam transverse cross section, (b-e) beam transverse cross-sections after passing through a linear polarizer whose polarization axis is indicated with a white arrow in the figure inset. (f) beam profile along the radial coordinate (comparison with theoretical prediction (dashed curve)).

Figure 2.15: (a) Picture of the commercial PbS QDs used in the experiment. The left is taken by a visible camera and the right is taken by an infrared camera. (b) Absorption curve of the QDs. (c) Emission curve of the QDs.

For the fluorescence collection properties of the fiber AXIGRIN, we design the experimental setup shown in Figure 2.16.
2.2. EXPERIMENTAL STUDY OF THE AXIGRIN PROPERTIES

We use a sample with infrared QDs embedded in a thin layer of PMMA A4. 50 µL original QDs dispersion as shown in Figure 2.15 (22.7 nmol/mL) was mixed with 50 µL PMMA A4 and then spin coated onto a cleaned microscopy cover glass with a spin speed of 5000 rps. The QDs are excited by focusing light from a laser diode at λ = 808 nm with an objective (20×, 0.4 NA). To ensure the focusing right at the QD layer, an additional objective is put on another side of the sample. With white light illumination, the QDs layer is imaged by a CCD camera with this latter objective. Then the position of the objective for laser focusing is adjusted to focus the laser onto the sample surface (QDs layer). If the laser is perfectly focused onto the sample surface, a tiny laser focal spot appears on the camera together with image of sample surface.

Because the numerical aperture of the objective for laser focusing is equal to 0.4, a spot size of about 2.6 µm is expected from conventional diffraction theory (Rayleigh criterion). Therefore, the QDs within an area of about 2.7 µm² can be excited, which can be seen as point fluorescence source compared to the AXIGRIN.

In the experiment as shown in 2.16, the AXIGRIN fiber system is fixed onto a piezo tube, which is controlled by a SNOM electronics from RHK company. RHK control module allows image acquisition in scanning microscopy, i.e. signal acquisition and transfer to a computer while the probe is raster scanned across the sample. The piezo tube is placed onto a 3D translation system (Thorlabs MAX350D/M), thus the distance between sample and AXIGRIN can be controlled. A photon counter from AUREA Technology is used to detect the in-fiber out-coupled signal that is collected by the AXIGRIN. Analog signal output from the photon counter is coupled into the RHK controller for image processing and stored with a computer. A long-pass filter (FEL1050, THORLAB) is used before the photon counter to stop the pump laser light at λ = 808 nm and make sure that what signal collected is the QDs fluorescence signal.

EXPERIMENTAL RESULT ANALYSIS

The fiber AXIGRIN probe is raster scanned across the single microscale fluorescent source at λ = 1500 nm to get the images. Due to defects in voltage amplifiers of RHK controller, we can just show images of limited area by 10 × 10 µm². The scanned images are shown in Figure 2.17(a). This figure is composed of two images respectively centered with respect to the fluorescent spot and at the bottom right area of the spot. It is clear that a bright spot is shown at the image center and there are concentric rings surrounding the central spot from the bottom right image. These rings are the signature of the "Bessel
response” of our probe in collection mode (given reciprocity principle). These acquisitions show a spot that is 3.7 µm wide (FWHM), which is wider than the theoretical predictions from diffraction theory (2.65 µm). This can be easily explained by the convolution effect of the fluorescent spot and the probe impulse response (Bessel function). Figure 2.17(b) shows the comparison between an image cross section along the white line in Figure 2.17(a) and the convolution between the typical Gaussian profile of the fluorescent spot response describing the AXIGRIN. The lateral peak positions are in good agreement. The amplitude reduce at the second lateral peak in experimental result is due to the collected signal decrease along the radial shift.

![Image](image.png)

Figure 2.17: (a) Scanning images of a micron-scale infrared fluorescence spot with a scan area of $10 \times 10 \mu m^2$ with sample-AXIGRIN distance at 1 mm. The bottom right image is the extended scanning image to show the surrounding ring. (b) Comparison of the cross-sectional curve (red) along the white line in (a) and a theoretical convolution curve (blue) of Gauss function and Bessel function.

Figure 2.18 shows images at various AXIGRIN-to-sample distances. Figure 2.18 (a), (b) and (c) are the images when this distance is 0 mm, 1 mm and 2.4 mm respectively. It is clear that the size of the central spot keeps almost unchanged in shape (it does not widen). This property is confirmed with the intensity profiles of Figure 2.18(d). The FWHM of the three spot images are measured to be about 3.7 µm. Figure 2.18(e) shows the evolution of the on-axis detected intensity as a function of the longitudinal propagation axis. We see that this on-axis detected intensity follows a function that is typical from Bessel-Gauss beams (see Figure 2.11(g)) [109], even if the AXIGRIN works here in collection mode (the AXIGRIN is not used to generate a Bessel beam but as a fluorescence collector). This shows that the detection volume of the fiber AXIGRIN probe is described by a Bessel function. Therefore, the main detection volume (of higher detection sensitivity for the AXIGRIN) takes the form of a sharp and ultra-long needle. The ultra-long field depth can reach as long as 3 mm as shown in 2.18(e).

**DIRECT FLUORESCENCE IMAGING WITH A CLEAVED OPTICAL FIBER**

As a comparison, we imaged the same infrared fluorescence spot without the AXIGRIN, i.e. directly with the bare cleaved optical fiber (SMF-28). In the experiment, the fiber is directly fixed to a 3D translation stage (Thorlab MAX350D/M) and manually moved along radial coordinates to scan the fluorescence spot step by step with one step of 4 µm. No automatic raster scanning is performed except for the preliminary centering process of the fluorescent spot within the image, before data acquisition. Figure 2.19(a) shows
2.2. EXPERIMENTAL STUDY OF THE AXIGRIN PROPERTIES

Figure 2.18: (a-c): scanning images of the microscale fluorescence spot with AXIGRIN- to-sample distance (z) of 0.2 mm (red curve), 1 mm (green curve), 2.4 mm (blue curve) respectively. (d) image profiles of (a-c) along radial coordinate. (e) on-axis profile of the detected fluorescence signal.

image profiles of the fluorescence spot, for three values of the fiber-to-sample spacing, z = 0 µm (red curve), z = 100 µm (green curve) and z = 220 µm (blue curve). Given the lower resolution ability of the fiber, the acquisition lateral extension has been widened to 16 µm, by realizing acquisitions with a manual translation stage. The FWHM significantly increases here with z: the initial value of 16 µm at z = 0 µm increases to 22 µm at z = 100 µm and 30 µm at z = 220 µm. We clearly see the added value of AXIGRIN in the fiber fluorescence imaging system, in term of depth of field and resolution ability. Figure 2.19 (b) shows the on-axis fluorescence signal as a function of the fiber-to-sample distance (z). This curve is obtained by moving the fiber along longitudinal direction z step by step with one step size of 0.1 mm. We see a drop of detected signal while z increases which confirms the very small collection depth of the bare cleaved fiber. With the AXIGRIN, the on-axis signal increases first with the increase of z up to 1 mm and then, it decreases for high z values which demonstrate that by adding the AXIGRIN, the collection depth of the fiber device is drastically enhanced.

AXIGRIN IN EXCITATION-COLLECTION MODE

We finally investigate the fiber AXIGRIN bench for fluorescence imaging in an all-fiber excitation-detection configuration that avoids the use of bulky optics. Figure 2.20 schemes the experimental set-up developed to demonstrate the fiber system. The spin coated QDs sample we prepared in the previous experiment is also used as the sample in this experiment. The pump laser diode (= 808 nm) is coupled to a 830 nm single mode fiber that is connected to an add-drop filter (ABSYS, 1 × 2 coupler, reflection band: 830 nm-870 nm, pass band: 1500 nm-1620 nm) aimed at separating the fluorescence signal at λ = 1500 nm from the pump signal at λ = 808 nm during the collection process. After the add-drop filter, the pump signal leaves the fiber, passes through AXIGRIN which generates the Bessel beam for exciting locally the sample. The AXIGRIN is also used to collect...
Figure 2.19: (a) Image profiles along radial coordinate with $z = 0\mu m$ (red curve), $z = 100\mu m$ (green curve) and $z = 220\mu m$ (blue curve), respectively; (b) on-axis fluorescence signal as a function of the longitudinal direction ($z$).

the fluorescence from the sample. The fluorescence is then out-coupled to the fiber and transmitted towards the detector (photon counting module from AUREA technology) by the add-drop filter (the pump signal is filtered out).

Figure 2.20: Experimental setup used to demonstrate infrared fluorescence imaging with fiber AXIGRIN in excitation-collection mode.

Figure 2.21 shows the detected fluorescence signal as a function of the AXIGRIN-to-sample distance. We need first to pay attention that due to the non perfect orthogonality between the sample surface and the probe axis, longitudinal translation of the probe is coupled to transverse slight shift. Therefore, it is really important to have homogeneously distributed QDs over the sample area to make sure that the measured changes of collected fluorescence signal are not due to variations of QDs concentration. The signal profile is fully consistent with the longitudinal signature of a Bessel beam, as shown in Figures 2.11(g) and 2.18(e). Here again, the signal increases with the increase of AXIGRIN-to-sample distance to maximum value at $z = 1\text{ mm}$, and then it exhibits a decrease for larger values of $z$. The little differences between Figures 2.18(e) and 2.21 can
be attributed to the fact that the noticeable spectral mismatch between the pump waves and the fluorescence signal. The chromatism of the AXIGRIN inevitably induces discrepancies between the Bessel beams generated at $\lambda = 808$ nm and $\lambda = 1500$ nm, in terms of Bessel beam length, residual divergence of the central spot, etc. However, it appears that these discrepancies are pretty small because the various longitudinal profiles measured at various wavelengths and in various AXIGRIN operation modes (excitation, collection and excitation-emission) are very similar. The chromatism of the AXIGRIN thus does not strongly impact imaging performances of the fiber probe.

![Figure 2.21: On-axis intensity change curve along longitudinal direction (z).](image)

To evaluate the imaging capability of the fiber AXIGRIN, in terms of extended depth-of-field, we processed the sample shown in Figure 2.22. QDs are spin-coated onto one of the two faces of two 170 µm thick microscope cover glasses. The other uncovered surfaces of the two glass plates are set in contact to a third microscope cover glass. The two QD-grafted glass plates are then shifted from each other to create a 1-millimeter wide gap without QDs. The different areas with and without QDs are thus separated by straight parallel edges and the two QDs layers are longitudinally separated by about 500 µm ($3 \times 170$ µm). The sample imaging is realized with the AXIGRIN working in excitation-detection mode.

![Figure 2.22: Scheme of double sides QDs sample.](image)

Figure 2.23(a) shows fluorescence plots with the fiber AXIGRIN probe moving along a
scan line perpendicular to the fluorescence edges of the sample (x direction), for three different AXIGRIN-to-sample distances given in the figure 2.23(a). The distance \( z \) is defined as the distance between the AXIGRIN and the front QDs sample as shown in Figure 2.22 so the distance between the AXIGRIN and the back QD sample should be 0.5 mm larger. In all three cases, the detected signal level from the 500 \( \mu \)m longitudinally separated QD layers is almost the same, which confirms the capability of Bessel beam to realize in-depth fluorescence imaging. The slight decrease of the collected signal from the back surface is mainly attributed to the scattering loss of the glass substrates. We see sharp detection peaks at the edges which are consistent with QDs accumulation at the glass plate edges during spin-coating process. Figure 2.23(b) shows the magnification of the dashed square in Figure 2.23(a) when \( z \) equals to 1 mm and the magnification curve shows the sample “edge response” width of 0.05 mm, which is much larger than the predicted imaging spatial resolution of 2.65 nm. The experiment/simulation mismatch is because the edge of the sample is not sharp enough.

![Graph](image)

Figure 2.23: (a) Fluorescence signal as a function of lateral shift (x) with three different AXIGRIN-to-sample spacing (z), blue, red and black curves for 1 mm, 0.75 mm and 0.5 mm spacing, respectively. (b) Magnification of the curve in the dashed square in (a) when \( z \) equals to 1 mm.

Therefore, the AXIGRIN appears to be a powerful fluorescence probe which enables ultrahigh depth of field fluorescence imaging with pump beam and fluorescence emission that are spectrally well-separated, which is a mandatory property for infrared fluorescence imaging, especially with with PbS QDs. In the case of PbS QDs, the pump beam has indeed to be spectrally located in the visible range and the fluorescence occurs at wavelengths about two fold larger. Such spectral mismatch is well beyond the chromatism range of conventional spherical optics. Thus, images shown in Figure 2.17 appear to be the first far-field infrared fluorescence images with PbS QDs. This system could therefore be of high interest in scientific, medical and industrial domain which pay more and more attention to infrared fluorescence imaging around telecommunication wavelengths.

### 2.3/ CONCLUSION

We introduced and demonstrated the new concept of fiber AXIGRIN probe which enabled the first infrared fluorescence imaging at telecommunication wavelengths (with PbS colloidal QDs). The fiber AXIGRIN probe is also compact, cheap, flexible and plug-and-play
2.3. CONCLUSION

which is of high importance for application point-of-view, especially for end-users that are non specialists in optics. The chapter includes the basic theory of Bessel beam generation, axicon and GRIN lenses. With the combination of axicons and GRIN lens, a new tool, AXIGRIN, was proposed and designed first to generate zeroth-order and first-order Bessel beams directly from a fiber output at $\lambda = 632$ nm. The AXIGRIN is integrated with an optical fiber aligned using a commercial pigtailed ferrule device. The properties of the Bessel beams are analyzed in detail and compared with theoretical predictions. The fabrication process of the new tool from commercial GRIN lenses is described in detail. In a second part, long depth-of-field fluorescence imaging on the millimeter range is demonstrated with a lateral spatial resolution on the micron scale. This structure is shown to overpass chromatism limitations, which unlock the full potential of far-field optics in infrared fluorescence imaging.

It also appears that the fiber AXIGRIN probe represents the first integrated confocal optical microscope including an axicon (to work in Bessel regime). Therefore, such a fiber probe provides a noticeable gain of resolution since the confocal configuration provides impulse response describe by $J^4$ instead of $J^2$ (to be demonstrated later).
3

BNA ANTENNA INTEGRATED FIBER SYSTEM

BNA (Bowtie Nano-aperture Antenna) has been intensively studied in optical regime due to its properties of intensity enhancement and super optical confinement when illuminated by light at resonance wavelength [117, 118]. It has been already applied in optical lithography [119, 120], optical tweezing [73, 121], fluorescence control [70] and nano-imaging. Bowtie was first proposed to be integrated at the end of an optical fiber to become a key interface between optical nano-objects and the fiber mode by Grosjean’s group [72], with direct applications in collection mode SNOM [122] (in-fiber near-field collection through the BNA) and fiber optical tweezers [73, 121] (BNA used in emission mode). Garcia Parajo’s group from ICFO successfully adopted this BNA on fiber tip as a SNOM probe in illumination/emission mode for high resolution single molecule imaging in visible spectral domain [79]. In the above cited works, BNA-on-fiber-tip was used either in illumination or in collection mode for SNOM application. In this chapter, we will propose a new BNA geometry that enables all-fiber imaging bench for fluorescence nano-imaging, down to single point-like emitters (such as QDs). During the single QD imaging process, the BNA fiber bench works in illumination-collection mode with no bulky optical lens adopted. The system is compact, flexible and easy to use.

In this chapter, we will first introduce SNOM since it is an important tool in our experimental study. Then the concept of BNA for SNOM nano-imaging will be introduced with a single resonance BNA aimed at near-field imaging photonic crystal Bloch modes (collection mode). Finally, the double resonance BNA will be introduced, theoretically investigated and experimentally validated for single infrared QD imaging.

3.1/ The use of SNOM

SNOM has become in the past years an important technique in nano-optics to "see" the nanoworld with a nanometer scale resolution down to a few nanometers [123], that is, far beyond the diffraction limit. It has been applied in many domains such as imaging [71], fluorescence imaging [78], sensing [75], photolithography [124], tip enhanced raman spectroscopy [125], nanoparticle tweezing [73] and manipulation [126]. In our research about near field fluorescence detection or imaging, SNOM is an important technique to use. So we will start with the introduction of SNOM systems.
3.1.1/ The Development of SNOM

The idea of SNOM was proposed as early as 1928 by Synge to overcome the diffraction limit in optical microscopy as shown in Figure 3.1 [127]. It was based on an opaque metal screen with a tiny subwavelength hole (A). The subwavelength hole is used to produce a nano source when illuminated with strong light behind the metal. A sample (S) is placed within a few nanometers in front of the subwavelength hole to be illuminated locally by the nano source. Images are then recorded point by point during subwavelength aperture raster scanning by detecting the scattered light from the sample. Due to the limitation of nano fabrication, the idea was not performed before 1984. With the development of high precision fabrication techniques, near-field imaging was experimentally demonstrated by Pohl's group [128]. In the new developed SNOM system, a nanoaperture worked in emission mode to illuminate samples in near field. An electronic feedback system was used to control the nanoaperture-to-sample distance into nanoscale. The sample scattered light was collected by optical objective.

In the modern SNOM systems, one important part is probe-to-sample distance control system. Many mechanisms have been proposed to realize it, such as electron tunneling [129], capacitance [130], photon tunneling [131] and near-field reflection [132]. But they are of limited versatility due to various disadvantages. For electron tunneling and capacitance control, conducting probes and sample surfaces are mandatory [133]. For the photon tunneling and near-field reflection control, limited bandwidth, difficulty in extracting topography information are their common problems [134]. Therefore, these techniques have been given up. In 1992, Betzig's group proposed a shear force feedback system to control the distance [134], which is an attractive choice with the advantages of versatility, high speed, low cost, and ease of use. The implementation of tuning forks for shear-force distance control has been suggested by Karrai's group in 1995 [135]. It has been widely applied in current SNOM systems. With this method, the probe is generally fitted onto a tuning fork which has high quality factor piezoelectric vibration resonances. When the probe is driven to approach sample at one resonance frequency, very small forces right at sample surface tend to slow down tip vibration, which leads to resonance shift of the tuning fork. This resonance shift is large enough to yield substantial changes in the amplitude and phase of oscillation. A feedback system is used to maintain amplification or phase of the tuning fork at setpoint value, which ensures distance control into the range of nanometer scale.
3.1. THE USE OF SNOM

Except the probe-to-sample distance control system, another important part of SNOM system is SNOM probe, which is the critical element to interact with samples in near field. Various probes have been proposed to be integrated directly with cantilever or tuning fork for near field scanning, such as tetrahedral tip [136], 90° prism [137], optical waveguide [138], fiber integrated probe [139] and so on. The probes can be used in four modes [133]: illumination mode, collection mode, reflection mode and perturbation mode. In illumination mode, the probe illuminates the sample in near field and the light from the sample is collected by bulky optics in far field [79]. In collection mode, the sample is illuminated in far field with dedicated bulky optics and the near-field information diffracted by the sample is collected by the probe in near field [132, 140]. In reflection mode, the probe is used both as light nano-source and as local light collector at the same time. Light from the probe illuminates the sample in near field and the same probe is used to collect near-field information from the sample [141]. In perturbation mode, the probe is neither used as nano-source nor as local detector. The probe is placed in near field region of sample to convert the near-field information of the sample into propagating far field light. Optical microscopy is used to illuminate the sample and collect the sample scattered light in far field [142]. In the various probes, one important probe is optical fiber based probe, which can be mounted onto tuning forks easily [139, 143]. Fibers with metal tip integrated [144], corrugated fiber tip [145], nanoantenna integrated fiber probe [79] have been reported with improved imaging resolution or energy output. In the fiber based probes, the nanoantenna integrated fiber probes, which combines the advantages of optical fibers and nanoantennas, have been intensively investigated. Monopole antenna [78], BNA antenna [79], gold elliptical antenna [146], etc have been developed to be integrated with optical fibers as SNOM probes with the applications of imaging [71], sensing [75] and so on.

3.1.2/ SNOM SYSTEM USED IN OUR EXPERIMENTS

The SNOM system in our experiments consists of three parts: SNOM head, SNOM electronics and computer. SNOM electronics and computer are used to control the SNOM head, thus control the probe position. We introduce here the information of SNOM head in detail. A commercialized SNOM head from NTMDT is used in the experiment. The SNOM head is placed onto an inverted Nikon microscope (Eclipse T E2000 – U) as shown in Figure 3.2. The sample (noted as “S”) is placed onto a microscopy holder. Conventional optical objective (OBJ) is used to illuminate the sample or observe the sample from a camera (not shown in the figure).

The SNOM head here consists of three parts: tuning fork, optical fiber tip and piezo electric tube based scanner. An optical fiber tip is glued onto the tuning fork (Farnell) by UV glue (Norway). The resonance frequency of the tuning fork is about 33 kHz. In our experiment, we glue the fiber not only onto tuning fork, but also onto a small plastic part to improve the stability as shown in the inset of Figure 3.2. The tuning fork/optical fiber system is fixed at one end of the piezoelectric tube based scanner. The scanner can be controlled to move the tuning fork/optical fiber system in the plane (X-Y plane) parallel to the sample surface to scan the sample with the maximum scanning area of 100 × 100 µm². A piezo electric tube is used here to approach the probe towards the sample in the direction (Z) perpendicular with the sample surface. A shear-force feedback system is applied to the piezo electric tube to control the probe-to-sample distance down to nanoscale. Therefore, the sample near field scanning can be realized with the piezo...
In the SNOM system, the SNOM head is supported by three legs (l₁, l₂, and l₃ as shown in Figure 3.2). Every leg height is controlled by the small screw at the top of the legs. The three legs are supported by holders which are denoted as h₁, h₂, and h₃, respectively. Holder h₁ and h₂ can translate the legs with micrometer resolution along x direction and y direction respectively, thus move the SNOM head in x-y plane. There is no translation screws in x-y plane for h₃. The three legs and holders can move roughly the fiber probe (with micron range precision) in 3D.

I practiced the above described experimental SNOM set-up involving BNA on tip in an experimental configuration simpler than the one involving single fluorescent emitters. The near-field images of a photonic crystal test-object with BNA fiber tip as local probe have been obtained. The sample is provided by Institut des nanotechnologies de Lyon in the frame of collaborative project with purpose of optical trapping [147]. The photonic crystal is composed of three different size periodic holes as shown in Figure 3.3(b). The surrounding periodic holes with radius of 112 nm are the largest-size holes. At the center, the holes with radius of 80 nm and 66 nm are fabricated in periodic way forming double period photonic crystal, which is a cavity that confines a Bloch mode in space and time. The double period is used to increase the cavity quality factor [148]. The photonic crystal works at the Γ-point of its band-diagram for direct addressing with a focused beam perpendicular to its surface. At resonance, the Bloch mode of the cavity efficiently confines the optical mode within the cavity, leading to a sharp dip (Fano resonance) in the reflection spectrum of the cavity. Since the photonic crystal is polarization selective, we define coordinates of x and y according to the photonic crystal orientation as shown in the insets of Figures 3.3(e) and (h). The corresponding reflection spectrum at the cavity of the photonic crystal for x direction polarized excitation shows the sharp dip at the wavelength of 1517 nm (Figure 3.3(c)). The experimental setup to observe the Fano...
onance of the photonic crystal is shown in Figure 3.3(a). Laser model 8164A (Hewlett Packard) which can offer the light within wavelength range of 1450 – 1620 nm is used. A long working distance objective (Nikon Mplan 60×, 0.7 NA, ELWD, 210/0) is used to focus the laser onto the sample to excite the photonic crystal. Since both the photonic crystal and BNA are polarization sensitive, it is necessary to provide the light with right polarization direction. So a half wavelength plate (λ/2 plate) is placed before the objective to modulate the excitation polarization. The reflected light is collected by the same long working distance objective and conducted into an optical spectrum analyzer at last, thus the reflection spectrum of the photonic crystal can be obtained.

Figure 3.3: (a) Experimental scheme for photonic crystal Fano resonance measurement. (b) SEM image of the photonic crystal. (c) Resonance spectrum of the photonic crystal with x direction polarized light excitation. (d) SEM image of the BNA. (e) and (f) Experimental and simulation images at the surface of the photonic crystal with BNA oriented along x axis, respectively. (see Figure (e) inset). (g) Image of the photonic crystal mode in the far-field zone with BNA oriented along x axis working in perturbation mode. (h) and (i) Experimental and simulation near field images at the surface of the photonic crystal with BNA oriented along y axis, respectively. (see Figure (h) inset). (j) Image of the photonic crystal mode in the far-field zone with BNA oriented along y axis working in perturbation mode.

For the SNOM imaging experiment, the BNA-on-tip (Figure 3.3(d)), which has been designed in the frame of Ali El Eter’s PhD (FEMTO-ST), is used as the SNOM probe for near field imaging. The BNA (260 nm BNA lateral width and 30 nm gap length) is fabricated onto a fiber tip apex coated with 100 nm thick Al film. With the BNA used in collection mode, the near-field information of the photonic crystal is detected by the fiber probe and conducted to an InGaAs detector through optical fiber guiding. When the BNA is oriented along x direction, the scanning image with the scan area of 10 × 10 μm² is obtained as shown in Figure (e). Then the sample is rotated by 90° so that the BNA is oriented along y direction and the near field image with the scan area of 10 × 10 μm² is obtained by the SNOM probe as shown in Figure 3.3(h). The images reveal two differ-
ent field distributions that are comparable with the simulation of the two perpendicular vectorial component of the electric field ($E_x$, $E_y$) parallel to the sample surface (Figures 3.3 (f) and (i)). This is consistent with the BNA polarization sensitivity in SNOM imaging, already shown in Refs. [72, 71, 122]. It reveals the electric dipole moment that drives the BNA at resonance. The SNOM set-up allows for simultaneous SNOM imaging in perturbation mode with field collection through the same objective as the one used for sample excitation ("OBJ" in Figure 3.3(a)). The images obtained for both BNA perpendicular orientations (Figures 3.3(g) and (j)) show that the photonic crystal resonance, which drives the sample reflectance, is affected by the presence of the tiny BNA. The subwavelength features detected in the far-field witness a near-field coupling between the two nanoscale structures. The noticeable modulation of the detected light during BNA raster scan across sample area reveal the resonant nature of the BNA. We have indeed a coupling between two resonators of strongly unbalanced quality factors which results in high perturbation of the resonator of higher quality factor, despite large mode volume, by the resonator of lower resonance, despite tiny mode volume. Perturbation of the photonic crystal with a non resonant nanoscale diffuser would not have a strong impact onto the photonic crystal resonance. This property confirms preceding works focused on new hybrid photonic plasmonic structures based on BNA [71, 149]. In the frame of this preliminary study, I have investigated and shown experimentally two important properties of the BNA on tip in a concrete experimental case.

2. / THEORY AND NUMERICAL SIMULATIONS OF BNAs

In the previous section, we showed qualitatively basic properties of BNA designed for photonic crystal near-field coupling. Here, we investigate BNAs properties theoretically and propose new BNA designs for nanoscale fluorescence mapping in ultra-compact all-fiber SNOM architectures that avoid bulky optics.

1. / FDTD METHOD FOR SIMULATIONS

FDTD (Finite Difference Time Domain) method is usually employed to realize the simulation of the BNA. FDTD method uses finite differences as approximations to both the spatial and temporal derivatives that appear in Maxwell’s equations [150]. Thus, space is meshed, on the basis of the Yee cell [150], and time is sampled for field calculation. All six facets of the computation volume are terminated with PML (Perfectly Matched Layer) to avoid unphysical parasitic reflection at the boundaries. All the parameters intrinsic to FDTD are widely described in many reports such as PhD manuscripts [151] and will not be detailed here. In the frame of my PhD, commercial FDTD code from SYNOPSIS (Fullwave) is used (https://optics.synopsys.com/rssoft/).

2. / CONFINEMENT PROCESS AND FIELD MODES IN A BNA

BNAs have the ability to enhance light confinements when illuminated at their resonance wavelengths. Such properties are due to a capacitive effect in between the two metal triangles, right at the gap, resulting in charge accumulation at optical frequencies and then light enhancement and confinement. This nanoscale physical effect is at the origin
of the electric dipolar properties of the BNA. It can be manifested with its high sensitivity to incident polarization: the highly asymmetric nanostructure generates optical hot spots when it is excited with incident polarization parallel to its metal triangles as shown in Figure 3.4(a). For perpendicular polarization, no field confinement is produced (see Figure 3.4(b)). This is a well-known behavior of dipolar antennas.

Figure 3.4: Experimental output spots of a BNA-on-tip fiber probe when illuminated by the incident light with polarization (a) parallel to and (b) perpendicular with the BNA metal triangles [72].

Such a physical process is related to the excitation of optical nanometer scale mode within the BNA. These tiny modes explain for instance that the resonance wavelength of the nano-antenna can be decided by the choice of metal and by its geometrical parameters [152]. BNAs are intrinsically multimode structures. The mechanism of the multi resonant modes inside BNA has been studied theoretically and experimentally [152, 153, 154]. Guo and co-workers [152] as well as Park and co-workers [153] attributed the multi resonant modes in BNA to the combination of plasmonic mode and Fabry-Perot like modes. The analysis from Ibrahim and co-workers proved that the multi resonant modes of the BNA are due to the combination of a guided mode and Fabry-Perot modes along the metal thickness [154]. We will explain the multi resonant modes according to this theory. Figure 3.5(a) in Ibrahim’s paper displays the modes of an infinitely long BNA, that is a BNA waveguide. The first mode is responsible for the capacitive effect at the origin of the field confinement. The second mode does not confine and enhance light and correspond to the mode excited at a BNA with the incident polarization perpendicular to the BNA axis that crosses the metal triangles. These two cross-polarized modes are waveguide modes and undergo dispersion curves. In reality, the BNA thickness is limited and behaves as a Fabry Perot that discretizes the dispersion curves of these modes: the propagative modes undergo multi-reflections at both BNA interfaces which over add geometrical resonances to each modes. Figure 3.5(b) displays transmission spectrum of a BNA as a function of the structure thickness. The BNA has geometrical parameters of 275 nm lateral width, 55 nm gap length and 800 nm thickness. The spectrum reveals the various Fabry-Perot resonances of the structure. The first resonant mode, noted $FP_0$ (fundamental Fabry-Perot mode), is excited at the waveguide cut-off and has thus an infinite phase velocity (near zero effective refractive index). Therefore, it exists whatever the BNA thickness is. We clearly see the zero effective index property of $FP_0$ whose spectral peak remains unshifted while varying the BNA thickness. The other Fabry-Perot modes show non-zero, varying, effective refractive indexes whose values are fixed by the Fabry-Perot effect (metal thickness).
The simulation results are shown in 3.5.

Figure 3.5: (a) Spectral density of the infinitely long bowtie waveguide made in gold with the BNA geometrical parameters of: 275 nm BNA lateral width and 55 nm gap length. (b) FDTD simulation result of the BNA transmission spectrum as a function of the metal layer thickness. The BNA parameters are same with the parameters in (a) with a thickness of 800 nm [154]

3.2.3/ Single resonance BNA, theoretical study

Usually, single mode BNAs are developed and only the first waveguide mode ($FP_0$) is used for applications. Spectral behaviors with perpendicular incident polarizations have been investigated [117]. Simulation results showed a much larger intensity enhancement with the incident polarization parallel to the long axis of antenna than that with perpendicular polarization. We first investigated this configuration for near-field fluorescence imaging with both fluorescence excitation and collection with the same nano-antenna on fiber tip (all-fiber nano-imaging platform).

The modeled configuration consists of a single resonance BNA at the apex of a metal-coated dielectric tip (Figure 3.6). The dielectric tip has a refractive index of 1.52 and a 500 nm radius-of-curvature rounded apex. The taper angle is about 14.3°. Taper angle and tip radius at apex are consistent with the angles and radius achieved with the photo-polymerization process of the tip fabrication [155]. Metal layer (Al) is 100 nm thick and covers the entire tip body. The geometrical parameters of the BNA are lateral size ($L$), gap length ($g$) and metal film thickness ($t$) as shown in the inset of Figure 3.6. The tip apex is positioned at a distance $d$ from a semi-infinite flat dielectric sample of refractive index of 1.52. BNA gap size is fixed at 40 nm in our simulation.

The FDTD computation volume spans 3.2 microns in X and Y about the apex of the tip. The apex is located at $X=Y=Z=0$, and the simulation spans 1 micron below the tip in the dielectric substrate and terminates 3 microns into the body of the tip. The length of the guiding part of the tip is about 3 micron long (1500 nm wavelengths) in our simulations. All six boundaries of the computation volume are terminated with perfectly matched layers in order to avoid parasitic unphysical reactions around the probe. The nonuniform grid resolution varies from 15 nm for portions at the periphery of the simulation to 5 nm in the
3.2. THEORY AND NUMERICAL SIMULATIONS OF BNAS

Figure 3.6: Scheme of the BNA-on-tip model.

region immediately around the apex ( spacings between tip and surface and at between metal tips at the BNA’s gap are described by 10 cells).

The BNA-on-tip is characterized spectrally in illumination mode. To this end, the waist of an input Gaussian beam ts the entrance aperture of the tip on which it is projected, at the upper part of the simulation. Emission spectra are shown for Gaussian beam in single temporal pulse regime. The time-varying electric field component parallel to the BNA axis $E_x$ is calculated at the middle of the metal layer insider the BNA gap (50 nm far from the BNA input and output end). The spectrum of the intensity of $E_x$ is calculated by a simple Fourier-transform of this result and normalized by the intensity spectrum of the free space propagating Gaussian beam, without the tip.

Figure 3.7: Simulation spectrums of single resonance BNA. The red, green and blue curves represent the resonance spectrums with BNA lateral width ($L$) of 280 nm, 275 nm and 270 nm, respectively. The solid and dashed curves represent the spectrums calculated with the substrate 10 nm and 15 nm away from the BNA, respectively. The BNA gap length ($g$) is fixed at 40 nm. The electric field is calculated at the center of the BNA gap and in the middle of the metal layer.

The resonance spectrums of BNAs with different BNA lateral width $L$ are shown in Figure 3.7. The red, green and blue curves are the resonance curves when $L$ is set at 280 nm, 275 nm and 270 nm, respectively. From the simulation results we can get that with
larger BNA size, the resonance wavelength is larger. The solid and dashed curves are the intensity spectrum with the dielectric substrate placed 10 nm and 15 nm away from the BNA, respectively. With the substrate placed farther from the BNA, the resonance wavelength is a little blue shifted. From the resonance spectrums between different BNA lateral size we can also see that the larger the BNA lateral size is, the larger resonance amplitude shows. There is no obvious difference of resonance amplitude with the dielectric substrate 10 nm and 15 nm away from the BNA.

Since the BNA is fabricated onto fiber polymer tip apex by FIB (Focus Ions Beam), the polymer over milling could occur during fabrication process. So it is necessary to evaluate the influence of the polymer milling on the resonance spectrum. The spectrum with and without taking polymer milling into consideration are shown in Figure 3.8. The blue, red, green and pink curves are obtained when the BNA lateral width are set at 210 nm, 240 nm, 270 nm and 280 nm, respectively. The solid and dashed curves represent the resonance spectrums with and without polymer milling. From the results we can see that the resonance wavelength with polymer milling is shorter than that without polymer milling. So with polymer etching, a blue shift of the BNA resonance wavelength would happen. There is also amplitude difference observed between polymer milling situation and non polymer milling situation. From the result we can also see that there is larger resonance amplitude with polymer milling.

![Figure 3.8: Simulation spectrums of single resonance BNAs with and without etching of polymer. The blue, red, green and pink curves represent the resonant spectrum when the BNA lateral width is set at 210 nm, 240 nm, 270 nm and 280 nm, respectively. The solid and dashed curves are the spectrums with and without polymer milling taken into consideration, respectively. The spectrums are calculated 15 nm (d) away from the BNA.](image)

From these curves one can estimate that a 40 nm large gap BNA with a lateral size $L$ of 280 nm is capable of resonantly interacting with the fluorescence emission from PbS QDs deposited onto a glass substrate (of emission wavelength around 1.5 microns) in SNOM configuration (tip to sample spacing of about 15 nm). This result takes realistic polymer over milling relative to FIB-fabrication.

Due to its near-zero effective refractive index, the fundamental mode $FP_0$ is responsible for high transmission efficiency of light [154]. It is thus capable of transmitting very efficiently fluorescence photons from single emitters positioned at one end of the BNA structure to the other end. This is consistent with SNOM fluorescence collection with
fiber BNA on tip (in-fiber fluorescence out-coupling) as already predicted theoretically [151]. In that configuration, the QD has to be in near-field coupling with the BNA, so it was positioned in close contact to the structure. An optical objective is employed to focus the light at 808 nm onto QDs sample to excite the QDs in far field. The BNA-on-tip was used just as the local detector to collect the QDs fluorescence. Due to the limited size of BNA, the transmission efficiency of the light at non resonance wavelength is very low. For the PbS QDs we use in our study, the excitation wavelength is located at 808 nm, which is far different from the QDs emission wavelength (1.5 micron). The mismatch between the two wavelengths makes it impossible for the single resonance BNA at 1500 nm to transmit the excitation light (808 nm) efficiently to excite the QDs. So bulky optical microscopy is not avoidable for QDs excitation with this single resonance BNA-on-tip probe detection. Developing an all-fiber system for QD excitation/detection is not realistic using the single resonance BNA.

3.2.4/ CONCEPT OF DOUBLE RESONANCE BNA FOR ALL-FIBER SNOM IMAGING

3.2.4.1/ DOUBLE RESONANCE BNA ON TIP

As an alternative to single resonance BNA for single PbS QD fluorescence nano-imaging, we propose the new concept of double resonance BNA compatible with all-fiber SNOM platform. This concept relies on the use of both the fundamental and first Fabry-Perot modes of the BNA. One resonance will be used to excite very locally the QDs (first Fabry-Perot Mode) and the other one to locally in-fiber collect the fluorescence emission (high transmittance fundamental mode). Given the unprecedented tunability of nano-antennas, the two resonances can be spectrally tuned independently from each other. This property is very favorable to "on-demand" implementation of SNOM tips for all-fiber fluorescence imaging, depending on the type of the fluorescent emitter envisioned.

We introduce and study numerically this new concept with a silver BNA, which can hold better double resonances compared to Al BNA. The simulation model is first based on a flat BNA milled inside silver layer with 200nm thickness. The double resonance spectrums of BNA with different parameters have been obtained and shown in Figure3.9. The gap size of the BNA is fixed at 30 nm. The red and green curves show the spectrums of the BNA with BNA lateral width of 270 nm, the BNA thickness of 200 nm and 150 nm, respectively. The blue and grey curves show the resonance spectra of the BNA with BNA lateral width of 240 nm, BNA thickness of 200 nm and 150 nm, respectively. The simulation result shows the influence of the geometrical parameters of the BNA onto its two resonant modes. We see that the variation of BNA thickness mainly tunes resonance wavelength of the first Fabry-Perot mode, with little impact onto the zeroth order mode (comparison between red and green curves and between gray and blue curves), whereas variation of BNA lateral width (L) tunes zeroth order Fabry Perot mode (see comparison between blue and red curves or gray and green curves). This unprecedented tuning capabilities of BNAs, originated by the guided mode nature of the metallic structure, allows very strong spectral mismatch between fluorescence excitation and collection, that can significantly exceed chromatism range of spherical optics used for the same purpose (objectives). Then fluorescence imaging of PbS QDs becomes possible.

In my PhD, I integrate the double resonance BNA at the end of a fiber-integrated metal-
coated SNOM tip in the purpose of single infrared PbS QD imaging. Given the extreme spectral mismatch requirements of PbS QDs between excitation (at 808 nm) and fluorescence detection (at telecommunication wavelengths), these quantum emitters represent a very attractive family of test-objects for unambiguously demonstrating our new "achromatic" all-fiber fluorescence nano-imaging concept.

In this study, the excitation wavelength is fixed at 808 nm and the fluorescence emission is centered to around 1500 nm. The BNA will thus have to develop double resonances at these two wavelengths. The simulation model is shown in Figure 3.10(a). The BNA is fabricated at the apex of a polymer tip ($x = y = z = 0$). The polymer tip radius is 500 nm with a silver film deposited. $L$, $g$ and $t$ represent BNA lateral width, gap length and metal thickness, respectively. A glass substrate with 20 nm thick PMMA layer deposited is placed 15 nm far away from the BNA. Numerical computation volume spans 6 microns in X and Y about the apex of the tip. The apex is located at X=Y=Z=0, and the simulation spans 1.5 microns below the tip in the substrate and terminates 5 microns into the body of the tip. All six boundaries of the computation volume are terminated with perfectly matched layers in order to avoid parasitic unphysical reactions around the probe. The nonuniform grid resolution varies from 15 nm for portions at the periphery of the simulation to 5 nm in the region immediately around the apex. With the BNA parameters of $L = 280$ nm, $t = 200$ nm, double resonance spectrums of the BNA with the gap length of 40 nm and 50 nm are shown in 3.10(b). The red and blue curves are for the BNAs with gap length of 50 nm and 40 nm, respectively. With the smaller gap size, the zeroth order resonance is a little red shifted. So the BNA lateral size and gap length can be modulated together to make sure
the the resonance wavelength is at 1500 nm. From the simulation result, we can see that the resonance wavelength of the blue curve is slightly larger than the required wavelength 1500 nm and the resonance wavelength of the red curve is a little smaller than 1500 nm, so in the further fabrication process, we can control the BNA gap size in between 40 nm and 50 nm.

Figure 3.10: (a) Simulation model. The polymer tip radius is 500 nm. L, g and t represent BNA lateral width, gap length and metal thickness, respectively. A glass substrate with 20 nm thick PMMA layer deposited is placed 15 nm far from the BNA. The BNA center is located at the position \( x = y = z = 0 \). With the silver BNA parameters of \( L = 280 \) nm, \( t = 200 \) nm, double resonance spectrums of the BNA within wavelength range of 0.5–2 \( \mu \)m is shown in (b) with the gap length of 40 nm (blue) and 50 nm (red), respectively. (c) and (d) are the electric field distributions inside the BNA gap (in x-y plane with \( y = 0 \)) along z direction at the wavelength of 800 nm and 1500 nm, respectively. (e) and (f) are the electric field distributions inside the PMMA layer (in x-y plane with \( y = 0 \)) along z direction at the wavelength of 800 nm and 1500 nm, respectively. (g) electric intensity profiles at both interfaces of the PMMA layer, at 800 nm and 1500 nm. Red and blue curves represent the distribution at the first interface (close to BNA) and the second interface (close to glass substrate), respectively. The solid curves and dashed curves represent the distribution at 808 nm and 1500 nm, respectively.

Figure 3.10(c) and (d) shows the electric field distribution inside the BNA gap (in x-y plane with \( y = 0 \)) along z direction at the wavelength of 800 nm and 1500 nm, respectively. Figure 3.10(e) and (f) are the electric field distributions right at PMMA layer at the two resonance wavelengths. From the Figures 3.10(d) we can see that for the electric field distribution along the BNA gap at 1500 nm, one maximum intensity spot turns up at the output end of the aperture and it is the BNA fundamental mode. Similarly, for the electric field distribution along the BNA gap at 808 nm, there are two bright spots and this corresponds to the first-order Fabry-Perot mode. From Figures 3.10(e) and (f) we can see that the light is well confined inside the PMMA layer. The electric intensity profiles at both interfaces of the PMMA layer at the two wavelengths are shown in (g). The red and blue curves represent
the electric intensity profile at the first interface (close to BNA) and the second interface (close to glass substrate), respectively. The solid curves and dashed curves represent the electric intensity profiles at 808 nm and 1500 nm, respectively. The spot sizes for the two wavelengths are almost same and at the second interface, the spot size is larger than that at the first interface (about 70 nm diameter at the first interface and 105 nm diameter at the second interface). The intensity decreases a lot from the first interface to the second interface despite 20 nm spacing due to the dispersion property of the PMMA layer. The electric intensity profiles of the spot in PMMA layer decides the imaging resolution.

3.2.4.2/ BNA-TO-QD COUPLING, INTRODUCTION OF THE INVOLVED QUANTUM PARAMETERS

To evaluate the imaging properties of the double resonance BNA in all-fiber architecture, we have to study the near-field coupling between the plasmonic structure on tip and a single quantum emitter. The basic parameters about single photon emission are introduced here. The parameters are decay rate, quantum yield and fluorescence enhancement. The excited state lifetime corresponds to the inverse of the decay rate.

DECAY RATE

During the spontaneous emission process of quantum emitters, energy is released in three forms. One is as an optical photon released into the external environment. This process is called radiative emission. The second possibility is to be absorbed by the external environment which is called nonradiative process. Thermal dissipation is one kind of nonradiative process. The third is internal energy loss of emitters associated with intramolecular dissipation, such as vibration loss and the energy will be released in the form of phonons [177]. The phenomenon is also called intrinsic loss. Total dissipated energy rate of quantum emitters is defined as decay rate \( \gamma \). We define \( \gamma_r, \gamma_{nr} \) and \( \gamma_i \) to denote the decay rate of radiative decay, nonradiative decay and intrinsic loss decay, respectively.

The relationship between total decay rate and the decay rate of radiative, nonradiative processes and intrinsic loss can be depicted as:

\[
\gamma = \gamma_r + \gamma_{nr} + \gamma_i
\]  \hspace{1cm} (3.1)

When emitters are put in homogeneous medium with no absorbance, the total decay rate \( \gamma \) can be written as:

\[
\gamma_{tot} = \gamma_0 + \gamma_i
\]  \hspace{1cm} (3.2)

where \( \gamma_0 \) is the radiative decay rate of the quantum emitters in a homogeneous medium and intrinsic loss \( \gamma_i \) in homogeneous medium is always constant. Surrounded by medium with no absorbance, there would be no nonradiative loss decay, so \( \gamma_{nr} \) should be 0.
QUANTUM YIELD

Quantum yield is defined as the probability of radiative emission of a quantum emitter when excited by external energy. It denotes the radiation efficiency of excited quantum emitters. Quantum yield η can be defined as:

\[ \eta = \frac{\gamma_r}{\gamma_r + \gamma_{nr} + \gamma_i} \]  

(3.3)

For the emitters placed in homogeneous medium without absorbance, the quantum yield can be depicted as:

\[ \eta_i = \frac{\gamma_0}{\gamma_0 + \gamma_i} \]  

(3.4)

where \( \eta \) and \( \eta_i \) are general quantum yield and intrinsic quantum yield, respectively. When the emitters are placed in homogeneous non-absorbing medium, in the case of \( \gamma_r = \gamma_0 \) and \( \gamma_{nr} = 0 \), general quantum yield \( \eta = \eta_i \).

LIFETIME

The excited states of an emitter are not stable and the emitters in excited states return to ground states as a consequence of radiative emission and nonradiative emission at last. Then, the lifetime of excited states can be defined as the time of an emitter being in excited states after excitation. The lifetime of an emitter placed in inhomogenous medium can be expressed as:

\[ \tau = \frac{1}{\gamma} = \frac{1}{\gamma_r + \gamma_{nr} + \gamma_i} \]  

(3.5)

while for the emitters placed in homogenous medium with no absorbance, lifetime can be depicted by the equation:

\[ \tau_0 = \frac{1}{\gamma_{tot0}} = \frac{1}{\gamma_0 + \gamma_i} \]  

(3.6)

\( \tau \) and \( \tau_0 \) are thus the excited state lifetimes when the emitter is placed in any inhomogenous medium and in homogenous medium without any absorbance, respectively.

To compare the spontaneous emission of an emitter in any environment and the emission of the same emitter placed in non-absorbing homogeneous medium, we define new parameters to depict the difference of the two situations: radiative decay rate increase factor \( (\Gamma_r) \) and total decay rate increase \( (\Gamma_{tot}) \). Radiative decay rate increase factor and total decay rate increase factor are defined by the equations as below:

\[ \Gamma_r = \frac{\gamma_r}{\gamma_0} \]  

(3.7)

\[ \Gamma_{tot} = \frac{\gamma}{\gamma_{tot0}} \]  

(3.8)
CHAPTER 3. BNA ANTENNA INTEGRATED FIBER SYSTEM

FLUORESCENCE ENHANCEMENT

Fluorescence enhancement is defined as the ratio of far-field fluorescence intensity from two identical emitters placed in two different environments. The two emitters in two different environments undergo the same excitation. The fluorescence enhancement can be depicted in two different forms according to excitation intensity. For one case, the fluorescence emission of an emitter in given environment (the quantum yield is fixed) just depends on excitation intensity. Its fluorescence increases linearly with the increase of excitation intensity until a critical intensity value \( I_c \) reaches. This process is called non-saturated regime. In the second case, excitation intensity is larger than \( I_c \), emitter fluorescence keeps constant and it doesn’t depend on excitation intensity. This process is called saturated regime. \( I_c \) is the boundary between the two excitation modes.

When emitters are excited in saturated regime, fluorescence enhancement \( \eta_F \) is equal to radiative decay rate enhancement \( \Gamma_r \) [156, 157]. The fluorescence in this mode no longer depends on excitation intensity, but affected (i.e. limited) by internal properties of emitters [156]. In the most commonly studied case, a structure (such as a nanoantenna) is placed close to an emitter in the first studied system and the other emitter is placed in homogeneous non-absorb medium. Then the fluorescence ratio between the two emitters can be expressed by the equation as below:

\[
\eta_F = \Gamma_r = \frac{\gamma_r}{\gamma_0} \tag{3.9}
\]

where \( \eta_F \) is the enhancement factor of saturation excitation.

When an emitter is excited in non-saturation mode, fluorescence enhancement is decided by the quantum yield of the studied system and the local excitation intensity at the position of emitters. The emitter fluorescence changes with the change of local excitation intensity. The second emitter is assumed to be placed in homogeneous medium with no absorption. To compare the fluorescence gain of the two identical emitters in two different environments, the fluorescence ratio of the two systems can be expressed by the equation below [26]:

\[
\eta_F = \frac{\eta \cdot I_e}{\eta_0 \cdot I_0} \tag{3.10}
\]

where \( \eta \) is the quantum yield of the studied system and \( \eta_0 \) is the intrinsic quantum yield of the emitter. \( I_e \) and \( I_0 \) are the excitation intensity at the position of the emitters in any environment and in homogeneous non-absorb environment, respectively. By replacing \( \eta \) and \( \eta_0 \) by equations 3.3, 3.4, we can get a new equation of the fluorescence enhancement \( \eta_F \):

\[
\eta_F = \frac{\gamma}{\gamma_0} \cdot \frac{I_e}{I_0} \tag{3.11}
\]

Then by using the equations 3.7, 3.8, the equation of fluorescence gain \( \eta_F \) can be rewritten as:

\[
\eta_F = \frac{\Gamma_r \cdot I_e}{\Gamma_{tot} \cdot I_0} \tag{3.12}
\]
The coupling of nano-antennas with quantum emitters is in the weak coupling regime, thus the quantum parameters describing the photon absorption and emission by the perturbed quantum emitters can be calculated from classical electrodynamics theory (probabilistic quantum description can be deduced from power flux calculation) [57]. Then the quantum description described above can be realized by FDTD method based on Maxwell’s equation, i.e. that considered light as electromagnetic wave and not by photons. The emitter is then modeled by a radiating dipole. However, this method is limited to calculation of decay rate enhancement since normalization is here mandatory to bridge classical and quantum electrodynamics [158]. This decay rate enhancement is known as golden Fermi’s rule and is fixed by the dipole source close environment [159].

We assume the far field radiations of emitters with and without special structure in close can be expressed as $P_r$ and $P_0$ respectively and the total energy emitted by the oscillating dipole (classical model of the emitter) with special structure in close is expressed as $P_{tot}$ ($P_{tot} = P_r + P_{nr}$, $P_{nr}$ is non radiative loss). $P_i$ is defined as the internal energy loss of the emitters. Then the total energy loss of the emitter with and without special structure in close are $P_{tot} + P_i$ and $P_0 + P_i$, respectively. The total decay rate enhancement $\gamma_{tot}$ and radiative decay rate enhancement $\gamma_r$ can be expressed as:

$$\gamma_{tot} = \frac{\gamma}{\gamma_{0tot}} = \frac{P_{tot} + P_i}{P_0 + P_i}$$  \hspace{1cm} (3.13)

$$\gamma_r = \frac{\gamma_r}{\gamma_0} = \frac{P_r}{P_0}$$  \hspace{1cm} (3.14)

where $\gamma$ and $\gamma_{0tot}$ represent the decay rate of the perturbed and free emitter, respectively. This equation is very important as it makes the connections between classical and quantum optics in our study.

Then, all the equation above mentioned can be extended to classical processing compatible with FDTD calculation with dipole emission source.

Note that emitters in homogenous medium without any absorption, $P_{tot} = P_r = P_0$.

Therefore, in saturation mode, the fluorescence enhancement can be expressed as:

$$\eta_F = \gamma_r = \frac{\gamma_r}{\gamma_0} = \frac{P_r}{P_0}$$  \hspace{1cm} (3.15)

Correspondingly, in non saturation mode, from equation 3.12, the fluorescence enhancement can be rewritten as:

$$\eta_F = \frac{\gamma_r I_e}{\gamma_{tot0} I_0} = \frac{\frac{P_r}{P_0} I_e}{\frac{P_{tot} + P_i}{P_0 + P_i} I_0}$$  \hspace{1cm} (3.16)

For the QDs with quantum yield of 0.4 in homogenous environment, that is $\gamma_r = 0.4 \times (\gamma_r + \gamma_i)$. $\gamma_i = 1.5 \times \gamma_r$, so $P_i = 1.5 \times P_0$, then we have:

$$\eta_F = \frac{\frac{P_r}{P_0}}{2.5 \times \gamma_0} + 0.6 I_0$$  \hspace{1cm} (3.17)
with the equation, the total quantum yield ($\eta$) of emitter can be expressed as:

$$\eta = \frac{P_2}{P_0^0} + \frac{1}{2.5}$$

(3.18)

Considering different emitter properties, such as single molecule, QDs and other diamond crystals, to use Born principle, we make the first approximation of emitters as electrical dipoles. The equation 3.16 can be written as:

$$\eta_F = \frac{P_2}{P_0^{tot}} \left( \frac{\eta}{2.5} \right) + 0.6 \left( \frac{|n_p \cdot E(r)|^2}{|n_p \cdot E_0(r)|^2} \right)$$

(3.19)

where $n_p$ is the unit vector which represents dipole orientation, $E(r)$ and $E_0(r)$ are the excitation electrical intensity at the position of the emitters with and without special structure in close.

The fluorescent molecules (chain-like) and quantum rods are nondegenerate linear emitters with 1D linear dipole emission and fixed polarization. The polarization influence to the fluorescence enhancement should be taken into consideration as shown in Eq. 3.19. For colloidal spherical QDs which have been demonstrated to be a twofold degenerate transition dipole oriented isotropically in two dimensions when excited by 2D polarized light [160, 161]. When excited by linearly polarized light, the emission is linearly polarized according to the excitation light polarization. So there is no polarization effect to the fluorescence enhancement.

### 3.2.4.3/ BNA-TO-QD COUPLING, THEORETICAL INVESTIGATION

The performances of the BNA-on-tip in all-fiber nano-imaging of dipole emitters is investigated by FDTD method with commercial “Fullwave” software. The configuration modeled here is shown in Figure 3.10(a). A 20 nm thick PMMA layer (considered as dispersive material, Fullwave library) lies on a glass substrate ($n=1.52$, non dispersive material). An emitter (dipole) whose dipole moment is oriented parallel to the interfaces is inserted into the PMMA layer, 10 nm far from the PMMA free surface. A metal coated tip is placed 15 nm far from the PMMA surface and centered with respect to the dipole.

Two configurations involving the same tip opened by two different apertures, a double resonance BNA and a conventional circular aperture, are considered. As noted in the previous section, the quantum description of the emitter-tip coupling can be realized with wave simulations from FDTD.

The parameter that will be measured experimentally is the fluorescence signal collected by the tip and guided to a detector. We will thus start our theoretical study with the calculation of the tip-collected fluorescence signal that can be expected with our setup. Tip-collected fluorescence enhancement calculated with the double resonance BNA will be normalized by the one achieved with the circular aperture, to position our concept with respect to a well-known and widely used SNOM nanoprobe. The design process of the on-tip double resonance BNA leads to $L = 280 \text{ nm}$, $g = 40 \text{ nm}$ and $t = 200 \text{ nm}$ with silver (not shown here, calculation of emission spectrum of the tip-integrated BNA, in “contact” to the sample, that fits our spectral requirements). The diameter of the circular aperture is
chosen to a value of 226 nm so that the opened areas of the circular and BNA apertures are equal. This allows for a direct comparison of the two apertures, in terms of excitation and collection efficiency which are parameters directly linked to the aperture area. Given the power injected into a SNOM tip, the fluorescence enhancements are calculated in non saturated regime.

As noted in Eq. 3.16 the fluorescence enhancement is the product of the excitation rate and quantum yield enhancements. The tip-collected fluorescence signal enhancement is then deduced as:

\[ n_{\text{tip}} = \frac{I_e}{I_0} \times q_{\text{tip}} \] (3.20)

where \( q_{\text{tip}} \) is the quantum yield into the tip, i.e. the probability that a photon emitted by the emitter is collected by the tip.

The calculation of the excitation rate for both tip apertures is realized on the basis of Figure 3.11. Here however, the intensity of \( E_z \) component (parallel to dipole orientation) is plotted at emitter’s position. The FDTD computation volume and gridding parameters are the same as for the previous FDTD simulations of the single and double resonance BNA on tip. The BNA-on-tip is characterized spectrally in illumination mode with the method already detailed for single resonance BNA simulation (see section 3.2.3). Figure 3.12(a) shows the ratio of excitation rate enhancements between the double resonance BNA and the circular aperture in a spectral domain ranging from 500 nm and 1000 nm. We see that the excitation rate with the double resonance BNA is about 100 times larger than with the circular aperture at \( \lambda = 808 \) nm.

The calculation of the quantum yield involves the replacement of the light source into the tip by a dipole source at the emitter’s position, oriented along (0x) axis. All the other simulation parameters stay unchanged, excepted the adding of field monitors to calculate the desired field components for power calculation. On the basis of the previous section, one can evaluate the quantum yield \( q_{\text{tip}} \) with:
where $P_{tip}$ is the power radiated by the dipole source into the tip and $P_{r}$ is the total radiated power. $P_{tip}$ is calculated from the Poynting vector flux through the tip cross section at the computation volume boundary. $P_{r}$ is achieved from the electric field component $E_{z}$ calculated during simulation at the dipole’s position (see Refs. [57, 151, 162]. FDTD simulations are conducted in pulse regime which allows for spectral studies of $q_{tip}$.

Figure 3.12(b) shows the spectrum of $q_{tip}$ achieved with the double resonance BNA, normalized by the spectrum of $q_{tip}$ with the circular aperture tip. Surprisingly, the curve that does not show any peak at $\lambda = 1500$ nm as one could expect with the zeroth order Fabry-Perot mode of the structure which is associated to high optical transmission. This means that the tip cutoff at its apex mainly controls the collection efficiency of the tip, the field radiated by the mode in far-field is not equally shared between the tip and free space. Cutoff phenomenon at the tip apex may prevent the full emission from the BNA to reach the tip body and drives (limits) tip collection. The curve of Figure 3.12(b) increases up to $\lambda = 1$ micron due to the absorption decrease of silver, and beyond this limit, it decreases due to cutoff phenomenon. We find however that $q_{tip}$, at $\lambda = 1500$ nm, is about 30 times larger with the double resonance BNA than with the circular aperture tip. And we have obtained that the excitation rate enhancement ratio between the two apertures of 100 as shown in Figure 3.12(a). Therefore, the detected fluorescence signal with the proposed all-fiber excitation/collection scheme is 3000 times higher with the double resonance BNA than with a circular aperture. A single emitter will be probed at 3000 photons per second with the double resonance BNA and at 1 photons per second with the circular aperture tip, at the same in-fiber input power for emitter excitation. Note that resolution of about 75 nm with the BNA would also fall down with the circular aperture of 226 nm diameter.

![Figure 3.12](image.png)

Figure 3.12: Excitation rate ratio between BNA and circular aperture. (b) Quantum yield $q_{tip}$ ratio between BNA and circular aperture.

In addition, since the BNA aperture and circular aperture are assumed to be placed in near field regime of the emitter, the emission decay rate will be influenced due to the near-field intervene of nanoapertures. In classic study, the total decay rate enhancement can be obtained using the Eq. 3.13. The total decay rate enhancements of the emitter with BNA or circular aperture in close to the free emitter in vacuum are analyzed and the results are shown in Figure 3.13. Figure 3.13(a) is the emitter total decay rate enhancement with BNA in close and (b) is that with circular aperture in close. With the influence of BNA, emitter total decay rate enhancement is about 105 times at 1500 nm wavelength, while for the circular aperture, the total decay rate enhancement reduces to 0.4 times. That means, the total decay rate with BNA aperture in close is 262 times of that with circular aperture in close. So the emission rate of the emitters coupled to the BNA is greatly
enhanced compared to circular aperture detection. The total decay enhancement of the BNA is consistent with the double resonance spectrum as shown in Figure 3.10(b).

Figure 3.13: (a) Emitter total decay rate enhancement with BNA in close. (b) Emitter total decay rate enhancement with circular aperture in close.

3.3/ **Experimental Study of Double Resonance BNA on Fiber Tip for All-Fiber Infrared Single QD Imaging**

The fabrication of the ultra-compact BNA fiber bench system is based on a commercial SMF – 28 optical fiber. The polymer tip with apex curvature radius of 500 nm is formed onto the cleaved end facet of the fiber by photopolymerization as depicted in Ref. [155]. Then 300 nm thick Ag film is deposited onto the fiber tip apex. After, 100 nm thick silver film at the very apex is removed by FIB (FEI Helios Nanolab 600i) and a flat surface with radius about 800 nm is left at the apex. At last, the BNA is milled into the silver film at the center of the fiber tip end surface by FIB. The SEM images of the fiber tips during the fabrication process are shown in the Figure 3.14.

Figure 3.14(a) shows the SEM image of the fiber tip after 300 nm Ag film deposited. The curvature radius of the tip is 500 nm. (b) is the fiber tip after 100 nm thick silver film being removed. (c) and (d) are SEM images of the fabricated BNA. The BNA lateral width is 285 nm and the gap length is about 47 nm, which are theoretical parameters as shown in Figure 3.10(b).

To realize single photon imaging, an experiment was designed as shown in Figure 3.15. Commercial colloidal QDs (Evidot, ED-P20-TOL-1500) dispersed in toluene with a concentration of 22.7 nmol/ml, which has been introduced in chapter 2 are employed here as quantum emitters. A laser diode (Thorlab LDC 205C laser diode controller+TCLDM9 diode mount+bluesky laser diode) of 808 nm wavelength is employed to excite the QDs. The laser light is collimated and passes through a short pass filter (FES0900, Thorlab, not shown in the scheme). The short pass filter is used to stop the long wavelength component noise in the laser. Then the excitation light is focused into an optical fiber (SM830, Thorlab), which is connected to a manual fiber polarization controller. The polarization controller is used to vary the laser polarization to adapt the laser polarization to BNA resonance polarization direction. Then the laser is conducted into an add-drop filter (AB- SYS, detailed information of the add-drop filter can be found in chapter 2), which is used to guide the excitation laser to the BNA-fiber bench. The BNA fiber bench is mounted onto a SNOM head and controlled by the SNOM system (see section 3.1.2). The BNA enhances the incident excitation light and confines it into nanoscale area. When the BNA
CHAPTER 3. BNA ANTENNA INTEGRATED FIBER SYSTEM

Figure 3.14: SEM images of the BNA-on-tip during fabrication process. (a) Image of the fiber tip with 500 nm curvature radius after 300 nm thick silver film deposition. (b) Tip after 100 nm thick silver film being removed by FIB. (c) BNA with the gap size of about 47 nm. (d) BNA from the end surface view. The BNA lateral size is about 285 nm.

is in contact to the sample, the single QDs distributed in the sample can be excited. Then the QD photons are collected by the BNA on tip as shown in the inset of Figure 3.15. The collected single photons are guided back into the add-drop filter, which can separate the QD fluorescence from the excitation laser. After passing through a U-bench where long pass filtering is carried out (with FEL1100, Thorlab), the QD fluorescence photons arrive at a photon counter (Aurea) to be detected and counted. The long pass filter is used to stop the excitation laser. The photon counter works in gate mode with a frequency of 62.5 kHz. The photon counter can convert the detected optical signal to analog electric signal that is transmitted to the SNOM signal port. During this image scanning, the SNOM software can record the BNA position together with the real-time fluorescence signal and therefore, the near-field scanning images can be obtained.

Samples with different QD concentrations are prepared for the near-field imaging. During the preparation process, we dilute the original QD dispersion with toluene to reach the desired QD concentration. Home-made PMMA solvent (PMMA A4, 4% weight PMMA powder dissolved in anisole) is used to mix with QD dispersion to form PMMA layer after spin coating. After 30 min shaking in ultrasonic machine, the mixture of QDs, toluene and PMMA solvent is spin coated onto a cleaned microscope cover glass with a spin speed of 2000 rps or 5000 rps. The thickness of the glass substrate is 0.17 mm which is compatible with confocal microscopy with immersion objectives. The PMMA layer thickness of the spin coated sample is mainly affected by PMMA content and spin speed. Six samples
Figure 3.15: (a) Experimental setup scheme for single QD imaging. The inset figure is the artistic view of the working principle of the double resonance BNA: QD excitation and single photon detection.

(S1-S6) are prepared with different QDs concentration, PMMA content and spin speed as shown in Table 3.1.

<table>
<thead>
<tr>
<th>parameters</th>
<th>S1</th>
<th>S2</th>
<th>S3</th>
<th>S4</th>
<th>S5</th>
<th>S6</th>
</tr>
</thead>
<tbody>
<tr>
<td>original QDs (µL)</td>
<td>30</td>
<td>25</td>
<td>10</td>
<td>10</td>
<td>2</td>
<td>1.5</td>
</tr>
<tr>
<td>toluene (µL)</td>
<td>20</td>
<td>25</td>
<td>80</td>
<td>80</td>
<td>85</td>
<td>85</td>
</tr>
<tr>
<td>PMMA A4 (µL)</td>
<td>50</td>
<td>50</td>
<td>15</td>
<td>20</td>
<td>15</td>
<td>15</td>
</tr>
<tr>
<td>Spin coating speed (bps)</td>
<td>5000</td>
<td>5000</td>
<td>2000</td>
<td>5000</td>
<td>2000</td>
<td>2000</td>
</tr>
<tr>
<td>Concentration (10^-7 mol/L)</td>
<td>68</td>
<td>56.7</td>
<td>22.7</td>
<td>22.7</td>
<td>4.54</td>
<td>3.4</td>
</tr>
</tbody>
</table>

PMMA layer thickness is measured for the sample S6 in Table 3.1. The sample was scratched carefully with knife to remove part of PMMA and the topography at edges is measured by AFM (Atomic Force Microscopy) in tapping mode. AFM image of the PMMA layer is shown in Figure 3.16(a). The black line in 3.16(a) is across the edge between PMMA area and PMMA-removed area. Figure 3.16(b) shows the topography along this line which reveals a thickness of about 20 nm, which is consistent with our simulation model. Notice that the height peak at the very edge is due to edge effect when we remove PMMA and it can not reflect the real PMMA layer thickness.

3.3.1/ Preliminary confocal optical images of QDs

As a preliminary optical characterization of the QDs, we tried to image our five PbS QD samples with home-made confocal microscope. This experiment is important because it would allow for first sample optical characterization before challenging SNOM imaging (to ensure for instance the presence of single QDs). This experiment would also be a test of the ability of confocal microscopy in infrared fluorescence imaging.

The imaging set-up is schemed in Figure 3.17.

The experiment is also based on the inverse optical microscope (Eclipse TE2000-U). Laser light at λ = 808 nm (from Bluesky Research laser diode) is used to excite the QDs sample. The laser light passes through a short pass filter (FES850, Thorlab) to remove
the long wavelength component noise. Then an oil immersion objective with 60× 1.2NA is employed to focus the light onto the QDs sample and detect the QDs fluorescence at the same time. The detected fluorescence is coupled into a multimode fiber with 50 micron core size (Thorlab M42L02, 0.22 NA, wavelength range: 400 – 2400 nm) by a lens and then conducted to a photon counter (Aurea) by the multimode fiber. A long pass filter (FEL1200, Thorlab) is used before the fiber to stop the short wavelength noise from the laser. The photon counter transform the detected photons into analog output, which is synchronized with the sample position information to obtain the confocal images. In the experiment, the sample is fixed onto a piezo plate, which is controlled by a controller from “RHK”. A software in this company can be used to control the piezo plate and scan the sample with the scan area of 4 × 4 µm².

Unfortunately, no images could be recorded with our confocal microscope. This is explained by the predicted chromatism of spherical optics which induces strong mismatch between the longitudinal positions of the objective focus at absorption and emission wavelengths (808 nm and 1500 nm in our case). PbS QD imaging is thus beyond spherical...
optics chromatism. Confocal microscopy remains unappropriated solution for PbS QD optical imaging. This could explain why we did not find any direct imaging of single QDs in the literature yet.

Therefore, to characterize our recipes in the generation of single QD samples, we decided to realize samples of visible QDs by following the recipes described above for PbS QDs. We chose CdSe/ZnS nanocrystals coated with octadecylamine ligands with a diameter of about 5 nm dispersed in toluene from MKnano company (MKN-CdSe/ZnS-T600) for this test. These QDs are characterized by absorption and emission wavelengths centered to $600 \pm 10$ nm and $620$ nm, respectively. The laser with wavelength of $532$ nm can also be used to excite the QDs, which is included within the chromatic correction range of usual objectives. Both the two kinds of QDs (visible and infrared) are colloidal nanoparticles of about the same size smaller that $10$ nm dispersed in toluene (PbS QDs are slightly larger). We thus make the hypothesis that the size and material discrepancy between the two particle families will not noticeably affect QD distribution within the sample after processing, provided that the QD concentrations into PMMA are almost identical.

The concentration of the product is $5$ mg/mL. For CdSe QDs with emission wavelength at $620$ nm, the CdSe core size is about $5$ nm and the average molar weight of the nanoparticles is $285000$ g/mol [163]. Based on the information we can obtain the molar concentration of the product of about $17.54$ nmol/mL. We prepared two samples with two different concentrations. For sample A, $10 \mu$L original QDs dispersion is mixed with $85$ $\mu$L toluene and $15$ $\mu$L PMMA A4, after 30mins shaking in ultrasonic machine, the mixture was spin coated onto a cleaned microscopy glass with a spin speed of $2000$ rps. For sample B, $2$ $\mu$L initial solution is mixed with $85$ $\mu$L toluene and $15$ $\mu$L PMMA A4 and is treated in the same way as the sample A. The QD concentration in sample B is similar to the one of sample S6 processed with PbS QDs. The QD concentration in sample A is in between the QD concentrations of samples S3 and S4 with PbS QDs.

To be adapted to PbS imaging set-up, the confocal architecture of the visible QDs imaging is similar to the one described above. $532$ nm He-Ne laser is used to excite the QDs. The laser light is collimated by an objective and passes through a short pass filter (FES550 Thorlab) and a polarizer. The polarizer is used to control the excitation intensity to $100$ $\mu$W level. The same objective as we used in PbS QDs experiment is also used here to focus the excitation light onto the QDs sample. The QDs fluorescence is then detected by the same objective and focused onto the entrance facet of a 50-micron diameter multimode optical fiber by an optical lens. The multi-mode fiber is also the same fiber as used in PbS experiment. Before the optical fiber, a long pass filter (FEL550 Thorlab) is used to stop the light from the laser. A visible photon detector (Aurea) is employed to detect the fluorescence photons conducted by the optical fiber. The photon detector transfers the optical signal to analog electric output, which is synchronized with the sample position information to obtain the confocal images. The images that we obtained with the two samples are shown in Figure 3.18.

Figures 3.18(a) and (b) are the confocal microscopy images of sample A and sample B, respectively. Figure 3.18(a) shows a distribution of spots larger than the diffraction limit imposed by the objective numerical aperture. The cross sectional plot along the white line of Figure 3.18(a) is displayed in Figure 3.18(c). It shows a FWHM of a single spot of about $570$ nm whereas diffraction limit is at $443$ nm in our case. Figure 3.18(b) displays a series of well defined and well separated spots of lower fluorescence signal level. These spots are smaller than in the case of sample A and show FWHMs at the diffraction limit,
Figure 3.18: (a) and (b) Confocal optical microscopy images of sample A and sample B, respectively. (c) and (d) Cross-sectional curves across the white line in (a) and (b), respectively. The scale bar of the images is 1µm.

as shown in the cross sectional plot of Figure 3.18(d) taken along the white line of Figure 3.18(b). These results tend to show that sample B provides well-separated single QDs.

This is confirmed by the blinking phenomenon observed in Figure 3.18(b), that is related to the imaging of single colloidal QDs. Blinking (emission on-off intermittency), which describes stochastic switching of fluorescence emission between photon emission mode (on mode) and nonradiative emission mode (off mode) under continuous excitation, is the competition result of emitter radiative emission and nonradiative emission[164]. Blinking is a unique property of fluorescent single emitters, such as single molecules and QDs. It is also an evidence to demonstrate single photon source used in the frame of this study.

Figure 3.19 shows time traces taken for a fixed position of the sample at a single spot of Figure 3.18(b), and for different excitation intensities. Blinking phenomenon is clearly observed. Figure3.19(a), (b) and (c) are the time traces with excitation powers $P_1 = 100 \mu W$, $P_2 = 60 \mu W$ and $P_3 = 4 \mu W$. It is clearly seen that the blinking phenomenon is related to excitation intensity. There are most 'off' states in (a) with higher intensity and least 'off' states in (c) with lower intensity. This result is in accordance with the blinking phenomenon shown in [165]. These time traces are perfectly repeatable in time on the same QD (see Figure 3.20). We obtained the same time traces and relationship with incident power for the other surrounding spots, which tends to show that sample B provides distributions of well separated single QDs. Therefore, sample S6 made with PbS QDs is highly desirable for our SNOM experiments in the demonstration of single QD imaging.

As a comparison, Figure 3.21(a) and (b) display time traces taken at a spot of sample A for incident powers $P_1 = 100 \mu W$ and $P_2 = 60 \mu W$, respectively. No blinking phenomenon can be observed in this case, which unambiguously proves that the larger spots observed in Figure 3.18(a) correspond to QD aggregates, or to single QDs very close from each
3.3. EXPERIMENTAL STUDY OF DOUBLE RESONANCE BNA ON FIBER TIP FOR ALL-FIBER INFRA

Figure 3.19: Blinking phenomenon of single QDs. (a) is time trace with excitation power of $P_1 = 100\mu\text{W}$; (b) is time trace with excitation power $P_2 = 60\mu\text{W}$ and (c) is time trace with excitation power $P_3 = 4\mu\text{W}$. We have: $P_1 > P_2 > P_3$

Figure 3.20: Blinking phenomenon of single QDs after certain time. (a) is time trace with excitation power of $P_1 = 100\mu\text{W}$; (b) is time trace with excitation power $P_2 = 60\mu\text{W}$ and (c) is time trace with excitation power $P_3 = 4\mu\text{W}$. We have: $P_1 > P_2 > P_3$
other (a few of them are located within the objective focal spot). This means that the other samples S1 to S5 made with PbS QDs show high QD densities and probably QD aggregates. These time traces are repeatable in time.

![Signal in infrared and SNOM photon QDs](image)

Figure 3.21: Detected signal time trace of the aggregates in Figure 3.18(a) with the excitation intensity of (a) $P_1 = 100 \mu W$ and (b) $P_2 = 60 \mu W$.

### 3.3.2 SNOM imaging of infrared QDs

The SNOM images of the samples S1 to S4 made with PbS QDs are shown in 3.22 (a), (c), (e) and (g), respectively, with various doubly resonant BNAs. The four images are obtained with four different BNA probes. As predicted by confocal images over visible QDs, the concentrations of the QDs are high enough to form QDs aggregates. The aggregates size gets smaller with the decrease of sample QDs concentration. The cross-sectional intensity curve of the QD aggregates along the white lines in the four images (Figure 3.22 ((a), (c), (e) and (g)) are shown in Figures (b), (d), (f) and (h), respectively. Note that our recipes lead to fluorescent samples, made of tiny nanoscale QDs clusters of homogeneous size across the sample surface that can be tuned with QD concentration. This type of sample could be used as an alternative to infrared fluorescent nanosize beads generally used as text-objects to characterize fluorescence imaging devices. Since fluorescent nano-beads do not exist with emission at telecommunication wavelengths, this type of sample would be highly desirable as they are very simple to fabricate. For instance, Figures (b), (d), (f) and (h) show edge responses of our double resonance BNAs on tips used in SNOM imaging. The 10%-90% resolution criterion of the BNA is estimated to about 75 nm (i.e. $\lambda_{\text{fluor}}/20$). This result is in the range of spot size and collection area of the doubly resonant BNA predicted theoretically from Figure 3.10(e) (in between 70 nm and 105 nm, within the PMMA layer, along longitudinal direction). The evaluation of the BNA resolution ability would be achieved also with single QDs. However, we relax here exigencies of collection efficiency since our fluorescent sources are brighter (due to higher QD concentration) and we have thus a first reliable estimation of the resolution of our all-fiber fluorescence SNOM in a less challenging configuration than the imaging of single PbS QDs. Figure 3.22(i) shows the time trace of the tip-collected fluorescence signal at an aggregate spot in Figure 3.22(a), no blinking is observed which confirms aggregate formation at the sample surface.

Signal levels of a few thousands counts per seconds are measured. Signal-to-noise ratio obtained here with photon counter detection is high enough to achieve non ambiguous images and first validation of our concept. The detected signal amplitude depends on
3.3. EXPERIMENTAL STUDY OF DOUBLE RESONANCE BNA ON FIBER TIP FOR ALL-FIBER INFRA

Figure 3.22: SNOM images of the S1 to S5. (a), (c), (e) and (g) are the near-field scanning images of the samples S1, S2, S3 and S4, respectively with the recipes as shown in table 3.1. (b), (d), (f) and (h) are the cross-sectional plots along the white lines shown in the SNOM images (a), (c), (e) and (g), respectively. The scale bar is 500 nm. (i) Signal time trace when the BNA is placed onto one spot of sample S1, no blinking is observed.
QD concentrations but also on BNA performances. We see in Figures (b), (d), (f) and (h) that the background signal is not at zero and is at different values on the different images. We explain this phenomenon by a residual fluorescence signal up to telecom- munication wavelengths of the polymer tip. It is excited with in-fiber pump photons and is back-reflected towards the detector. Then, the signal from the QDs is over added to this background signal. This background signal is directly proportional to the input power. It thus reflects either the BNA capability to concentrate light in the near-field or the de- gree of alignment of the incident polarization (linear) to the BNA polarization axis, which drives resonance excitation. Measuring noticeable fluorescence signal with low back- ground means that the resonant enhancement of light with the BNA is efficient and/or that the impinging linearly polarized fiber mode is aligned with the metal triangles of the BNA. In contrary, a lower light enhancement efficiency and/or polarization mismatch with BNA mode would involve higher incident power to collect enough fluorescence signal, and thus higher background signal from the tip for the same signal level. From the above considerations, a better configuration is met with sample S5 imaging as shown in Figure 3.23 (a) and (b) since the background signal is the lower one with a signal dynamic of about 1700 cts/sec for almost single photon source mapping. This background signal has no consequence onto the SNOM images, a constant signal level is added to the fluo- rescence signal of interest. However, it will prevent us from measuring QD fluorescence decay as well as demonstrating single photon emission by measuring second order auto- correlation function [166].

![Figure 3.23](image)

Figure 3.23: (a) SNOM images of sample S5 as shown in table 3.1. The scale bar is 500 nm. (b) Cross-sectional plot along the white line in (a)

QD concentration of S5 is about 1.4 higher than the one of sample B made with visible QDs which led to well separated single QDs. The spot size (FWHM) is here measured to 90 nm which is slightly larger than the 10%-90% resolution criterion measured on the same image. This tends to prove tiny aggregates of a few QDs. Probably single QDs are present but there will be always a doubt in the nature of the observed light spots. We thus further diluted the QDs to reach QD concentration similar to sample B and then to expect well separated single QDs. We mixed 1.5 µL original QD dispersion with 85 µL toluene and 15 µL PMMA A4 solvent to get the mixture with QDs concentration of about 0.3 nmol/mL. After 30 mins shaking in ultrasonic machine, the QDs were spin coated onto a cleaned microscope cover glass with a spin speed of 2000r ps. This sample is called S6 in table 3.1 which summarizes sample recipes.

Figure 3.24 shows SNOM images with our all-fiber system involving two doubly resonant BNAs (the experimental set-up stays unchanged, see Figure 3.15). Figures 3.24(a) and
3.3. EXPERIMENTAL STUDY OF DOUBLE RESONANCE BNA ON FIBER TIP FOR ALL-FIBER INFRA

(b) are achieved with the first BNA while Figures 3.24(c), (d) and (e) are obtained with the second one. Figures 3.24(d) is a zoom of Figures 3.24(c). Figures 3.24(e) is the image of another area of the same sample with a scan area of $1 \times 1 \mu m^2$.

There is slight difference in detected signal between the two BNAs. The two series of images show deeply subwavelength spots of uniform size and of varying intensities (the scan area of Figures 3.24(a) and (b) is one fluorescence wavelength). Intensity variations are attributed to two possible reasons: the height difference between various QDs within PMMA layer and the different intrinsic quantum yields of the QDs. However, the spots have almost the same pattern which is consistent with their particular dipolar emission properties [160].

Spot size in the images obtained with the first BNA can be deduced to values about 75 nm (FWHM), i.e. $\lambda f_{luo}/20$, from the image profile shown in Figure 3.24(f) (image profile along the white line of Figure 3.24(b)). With the second BNA, the spot size is estimated to about 70 nm (FWHM), i.e. $\lambda f_{luo}/21.5$, from the image cross section shown in Figure 3.24(g) (image profile along the white line of Figure 3.24(d)). These results are repeatable over the various spots of all the images of Figure 3.24. They are fully consistent with the light confinements numerically predicted to be produced by the double resonance BNA within the PMMA layer, at wavelengths of 808 nm and 1500 nm (in between 70 nm and 105 nm along z-direction, see Figure 3.10(e) and (f)). Note that the confinement produced at 1500 nm (Figure 3.10(f)) is supposed to define the detection volume of the nano-antenna in collection mode for our infrared fluorescence imaging application.

Figure 3.24(h) shows the detected signal time trace when the BNA is located at one spot position in Figure (b). In contrary to the time trace of S5, blinking phenomenon is clearly observed here, which is the demonstration of single photon detection from single QD. Since all the spots of the images have uniform size and of the same signal level, one can assert that well separated single QD are dispersed into the thin PMMA layer, as first shown for sample B (see Figure 3.18(b)) with the same QD dilution. We have thus the experimental demonstration of the ability of our concept of double resonance BNA to form ultra-compact nano-optical platforms for all-fiber fluorescence nano-imaging down to single QDs. Figure 3.24 represents to our knowledge the first images of single PbS QDs. We therefore demonstrate fluorescence imaging over ultra wide spectral range well beyond objective operation bandwidth.

Figure 3.25 shows the BNA signature in the imaging process, by emphasizing its polarization sensitivity. Figure 3.25 (a) and (c) show the simulation of the electric intensity produced by the doubly resonant BNA-on-tip at excitation wavelength, for two orthogonal orientations of the incident electric field, parallel and perpendicular to the BNA’s metal triangles, respectively. In-fiber BNA excitation at $\lambda = 808$ nm is considered (see Figure 3.10) and the intensity is plotted at the middle of the PMMA layer in x-y plane (i.e. $10nm$ far from the layer upper interface and $25nm$ away from the BNA). The simulation results (Figures 3.25(a) and (c)) are compared to the experimental images of single QDs (sample 6)(Figures 3.25(b) and (d), respectively) that involve the same incident polarization conditions as in simulations. The incident fiber mode impinging onto the BNA (at 808 nm) is linearly polarized along the BNA’s metal triangles (Figure 3.25(b)) and perpendicularly to this nano-antenna axis (Figure 3.25(d)). For incident polarization parallel to the BNA axis (Figure 3.25 (a) and (b)), simulation shows a highly localized electric field enhancement right at the BNA’s gap, which was previously described as the starting point of the nano-imaging process (spot size of about 75 nm). The experimental image show two
CHAPTER 3. BNA ANTENNA INTEGRATED FIBER SYSTEM

Figure 3.24: SNOM image of sample 6 (low concentration sample). (a) and (b) Images with a scan area of 1.5µm × 1.5µm and with the first BNA on fiber tip. (c), (d) and (e) are near field images with the second BNA. Their scanning areas are 1.8 µm, 500 nm and 1 µm, respectively. The scale bar of images (a), (b) and (c) is 500 nm. The scale bars of (d) and (e) are 125 and 250 nm, respectively. (f) and (g) Cross-sectional intensity change along the white line in (b) and (d), respectively. (h) Detected signal time trace at a single spot. Blinking phenomenon is observed which proves single photon emission from single QD.

well defined spots whose FWHMs are about 75 nm. When the incident polarization is turned by 90° (Figure 3.25 (a) and (b)), the bright spot initially found right at the gap in the simulation disappears and the electric intensity level is much lower. The corresponding SNOM image does not exhibit well-defined spots anymore at QD positions and weaker fluorescence signal is detected. Such imaging behavior of our SNOM confirms that the BNA is responsible for the imaging process.

In addition, the fiber tip shape is observed to be a factor affecting QD emission/detection
3.4. **CONCLUSION**

In this chapter, we developed an all-fiber system with double resonance BNA integrated for single infrared QD imaging. Since SNOM is an important technique in our research, the principle and the components of SNOM system was first introduced in detail. The performances of the system. The comparison between two kinds of tips is shown in Figure 3.26. A well defined conical shape of the tip body (Figure 3.26(a)) led to the higher detected fluorescence signals. Tips with cylindrical tubular shapes, as shown in Figure 3.26(b), detected lower signals from fluorescent samples. Therefore, the tip shape appears to strongly affect the imaging performances of the SNOM. This experimental consideration is in agreement with the simulation result of Figure 3.13(b) which brought the conclusion that the collection efficiency of the BNA on tip is (surprisingly) mainly driven by the tip properties instead of BNA resonance (Fabry Perot zeroth mode). The tip shape and size are thus crucial parameters that need to be optimized. There is thus a need to fabricate the fiber tips with proper shape properties and this point needs further investigations.

![Figure 3.25: Simulation of the electric intensity at 800 nm with the incident light polarization parallel (a) and perpendicular (c) to the BNA's metal triangles, respectively. (b) and (d) are near field scanning images of infrared QDs with the incident light polarization parallel and perpendicular to the BNA's metal triangles, respectively.](image)

![Figure 3.26: SEM images of two BNA fiber tips with two different shapes. (a) BNA SEM image with higher experimental performances. (b) BNA SEM image with lower experimental performances. The scale bar is 1 µm.](image)
concept of BNA for SNOM nano-imaging will be introduced with a single resonance BNA aimed at near-field imaging photonic crystal Bloch modes (collection mode). Then a double resonance BNA-fiber bench was designed with the BNA milled inside Ag film. Theoretical basis of single quantum emitter and the efficient coupling between single quantum emitter and double resonance BNA were presented. Single photo imaging for the infrared QDs at telecom wavelength with the all fiber system has been experimentally demonstrated. The excitation of the QDs and QDs fluorescence detection were realized at the same time with the fiber system. The near-field scanning images from high concentration sample and low concentration sample are obtained. The influence of excitation polarization to the imaging quality is studied and the high resolution as high as $\lambda_{flu}/20$ has been obtained. Blinking phenomenon has been observed to prove the single QD distribution of the sample. The most important property of the double resonance BNA is that it can be applied within a broadband optical range. All the illumination-detection systems are based on optical fibers with the advantages of compact, plug-and-play and feasible.
Fiber integrated nano-optical horn antenna for in-fiber luminescence collection

Horn antenna concept is a simple alternative to the BNA for in-fiber luminescence detection. It has the advantage of high emission directivity with dipolar point-like source, which is needed for fluorescence fiber collection with tiny structures. In this chapter, we investigate the concept of nano-optical horn antenna for in-fiber luminescence out-coupling, with the exploration of fiber integrated "on-demand" single photon sources and X-ray excited luminescence detection.

In this chapter, we will first introduce the basic theory of the horn antenna, the extension of this antenna concept to nano-optics for controlling light dipolar sources and quantum emitters. The fabrication process will be depicted in the second section. In the third section, we will apply horn antenna to QDs infrared fluorescence coupling experiment. Finally, nano-optical horn antenna will be applied to X-ray excited optical luminescence in-fiber collection. In this application, the resulting ultra-compact horn antenna-fiber platform can be further exploited as an X-ray sensor.

4.1 Horn antenna for dipole emitter coupling into optical fiber

4.1.1 Horn antenna concept in microwave regime

In microwave regime, horn antenna consists of a tapered metallic hollow waveguide, metallic mirror and coaxial cable [167] (Figure 4.1). It flares into an open ended conical or pyramidal shaped horn. It transforms the impedance of a monomode metallic hollow waveguide (high impedance) to the impedance of free space of 377 \(\Omega\) (lower impedance) simply by flaring the waveguide into a larger opening. Usually the horn antenna is coupled to coaxial cable with a coax-to-waveguide transition [168]. The overall transition+horn structure can couple the end of the coaxial cable (i.e. a dipolar source), into free space and it leads to highly directional emissions and collections.

In this architecture, the coaxial cable end wire, which radiates as microwave dipole, is placed in between the metal reflector of the coupler and the tapered waveguide with a
distance of \( \lambda_g/4 \) from the reflector, where the \( \lambda_g \) is the effective wavelength of the waveguide mode. The distance is chosen according to constructive interference principle. When the distance is \( \lambda_g/4 \), the microwave signal is constructively interference. The distance between the reflector and the wave source is carefully chosen from analysis of impedance matching between the coax and the waveguide to make the back reflected radiation from reflector interferes constructively with the radiation that propagates directly toward the opening, which leads to a high-efficiency radiation directed outside the waveguide.

Based on the concept of horn antenna in microwave regime, a high-efficiency collection optical horn antenna, which is directly integrated with a single mode optical fiber for efficient single dipole emitter coupling, has been proposed theoretically by Grosjean et al [169].

4.1.2/ Concept of Fiber Nano-Optical Horn Antenna and Theoretical Study

The Configuration of Optical Horn Antenna

The scheme of the proposed optical antenna is shown in Figure 4.2.

The nano-optical horn antenna is directly adapted from the overall microwave structure "horn+transition" introduced above. It consists of a tapered dielectric tip and a gold mirror, corresponding to the flaring waveguide of the horn antenna and the reflector of the transition, respectively. An emitter with emission dipole moment oriented perpendicular to the optical axis of the horn antenna, is put at the very tip of the dielectric horn, with the mirror placed in front of the tip at a distance of \( \lambda/4 \). The horn antenna is designed to achieve directive emission from the nanoscale light sources which efficiently match the impedance of single mode fiber. The horn antenna can be easily integrated with a cleaved fiber by being placed in contact with the cleaved end facet of the optical fiber (also called \( \pi \)-plane).
4.1. HORN ANTENNA FOR DIPOLE EMITTER COUPLING INTO OPTICAL FIBER

Figure 4.2: Scheme of the fiber connected optical horn antenna [169].

if the horn antenna refractive index is chosen close to the fiber core refractive index.

NUMERICAL SIMULATION OF HORN ANTENNA FOR DIPOLAR EMITTER COUPLING

The simulation scheme of the horn antenna-fiber system is shown in Figure 4.2. A single mode optical fiber ($SMF - 28$) at telecommunication wavelength with core diameter of 8.2 µm is considered. The refractive index of fiber core and cladding are 1.46 and 1.45, respectively which are given in the fiber data sheet. The curvature of the horn antenna apex is expressed by $R$. The distance between mirror and horn antenna apex and horn antenna base diameter are depicted as $h$ and $D$, respectively. The collection efficiency and the directivity of the horn antenna are studied theoretically.

The collection efficiency of the horn antenna-fiber system, that is the ratio of the photons coupled from the emitter to the optical fiber can be expressed by the coefficient $T$:

$$T = q_{tip}C_m$$  \hspace{1cm} (4.1)

where $q$ is the quantum yield of the emitter, $q_{tip}$ is the quantum yield into the fiber tip, that is the collection efficiency of the horn antenna as defined in equation 3.21 and $C_m$ is the coupling efficiency of photons from horn antenna to the single mode optical fiber. Factor $T$ represents the ratio of emitted photons that are collected by the horn antenna and outcoupled into the fiber mode. Quantum yield $q$ is defined as:

$$q = \frac{\gamma_r}{\gamma_r + \gamma_{nr}}$$  \hspace{1cm} (4.2)

where $\gamma_r$ and $\gamma_{nr}$ are the radiative and non radiative decay rates of the emitter, respectively. The emitter is assumed to be an ideal emitter with the intrinsic loss as 0, that is $\gamma_i = 0$. $\gamma_r$ is defined as: $\gamma_r = P_r/P_0$ and $\gamma_{nr} = P_{nr}/P_0$ where $P_r$ is the far field radiation from the emitter coupled to the horn antenna, $P_{nr}$ is the emission loss dissipated by the horn antenna and $P_0$ is the radiated intensity by the emitter in free space without the presence of the antenna.

The collection efficiency $q_{tip}$ of the horn antenna is defined as the fraction of radiated power that is channeled within the body of the flaring dielectric waveguide. It is obtained from:
where $P_{tip}$ is the power collected and conducted by the horn antenna.

Coupling efficiency between the horn antenna and the optical fiber can be expressed in terms of the coupling coefficient $C_m$, defined as:

$$C_m = \frac{P_m}{P_{tip}}$$

where $P_m$ is the emitter radiation coupled into the single mode fiber. $P_m$ and $P_{tip}$ are obtained from Poynting vector integrations over the calculated area in optical fiber entrance facet.

\[
P_i = \frac{1}{2} \Re e \int_0^\infty \int_0^{\pi/2} r^2 e^{jkr \sin \theta} E^*_i \times H^*_t \cdot \hat{e}_z r r d\theta d r
\]

\[
P_m = \frac{1}{2} \Re e \int_0^\infty \int_0^{\pi/2} r^2 e^{jkr \sin \theta} (a_m E^*_m \times b^*_m H^*_m) \cdot \hat{e}_z r r d\theta d r
\]

where the unit vector $\hat{e}_z$ defines the direction of the fiber axis and the expressions of constants $a_m$ and $b_m$ are based on the overlap integrals between the collected electromagnetic fields output from the horn antenna ($\hat{H}$) and the fiber mode field distribution ($\hat{E}$).

\[
a_m = \left\{ \begin{array}{l} \frac{\int_0^\infty \int_0^{\pi/2} E_i \times H^*_t \cdot \hat{e}_z r r d\theta d r}{\int_0^\infty \int_0^{\pi/2} E^*_m \times H^*_t \cdot \hat{e}_z r r d\theta d r} \\ \frac{\int_0^\infty \int_0^{\pi/2} E^*_m \times H^*_t \cdot \hat{e}_z r r d\theta d r}{\int_0^\infty \int_0^{\pi/2} E^*_m \times H^*_t \cdot \hat{e}_z r r d\theta d r} \end{array} \right.
\]

\[
b_m = \left\{ \begin{array}{l} \frac{\int_0^\infty \int_0^{\pi/2} E^*_i \times H^*_t \cdot \hat{e}_z r r d\theta d r}{\int_0^\infty \int_0^{\pi/2} E^*_m \times H^*_t \cdot \hat{e}_z r r d\theta d r} \\ \frac{\int_0^\infty \int_0^{\pi/2} E^*_m \times H^*_t \cdot \hat{e}_z r r d\theta d r}{\int_0^\infty \int_0^{\pi/2} E^*_m \times H^*_t \cdot \hat{e}_z r r d\theta d r} \end{array} \right.
\]

Another critical parameter that describes the performances of optical horn antenna (and antennas in general) is the directivity:

$$\text{Dir} = \max \left( \frac{4\pi P(\beta, \theta)}{P_r} \right)$$

where $P(\beta, \theta)$ is the angular power radiated in the direction of polar angle $\beta$ and azimuthal angle $\theta$, and $P_r = \int P(\beta, \theta) \sin \beta d\beta d\theta$ is the integral over all angles. An isotropic source would have a directivity of 1, whereas for a dipolar emitter $\text{Dir} = 1.5$.

**Simulation results**

Numerical simulations are realized with 2.5 FDTD method [170], the simulation termi- nates 42 microns into the body of the tip, that is at the end facet of the dielectric horn.
4.1. **HORN ANTENNA FOR DIPOLE EMITTER COUPLING INTO OPTICAL FIBER** 85

The nonuniform grid resolution varies from 15 nm for portions at the periphery of the simulation to 5 nm in the region immediately around the apex. For accurate simulation of the horn antenna optical properties, the lateral extent of the FDTD computation volume was chosen to be wide enough (15 µm along the radial coordinate) that the captured field distributions were significantly wider than the lateral extent of the fiber core. Since the computation volume terminates at the end facet of the dielectric horn, the fields calculated at the end facet can be directly used for the calculation of the mode coupling into an optical fiber. The horn antenna to fiber coupling is simulated with overlap integral method [171].

In the simulation, the dipole emitter with radiation at a wavelength of 1.45 µm and two horn antennas with different geometries are considered. One antenna is designed with the tip radius curvature (R) of 0.75 µm, the horn antenna base diameter D of 12 µm and the antenna-mirror distance (h) of 0.39 µm. The other horn antenna is designed with the parameters of R = 0.5 µm, D = 10 µm and h = 0.29 µm. The most important parameters to evaluate horn antenna collection efficiency and optical horn antenna directivity. The collection efficiency is described as the ratio of the power transmitted inside the circular area of radius r contained in the τ-plane as shown in Figure 4.2. The simulation results of the collection efficiency of the two horn antennas are shown with the blue curve and red curve, respectively, in Figure 4.3(a). The two vertical dotted lines represent the lateral limit in the τ-plane of the two horn antennas considered here. As a comparison, the power ratio achieved in the τ-plane without any horn antenna is also calculated and shown by the green curve in Figure 4.3(a). The power achieved in the τ-plane without any horn antenna is very low as shown by the green curve. From the simulation result we can see that the maximum collection efficiencies of the two antennas are 80% and 85%, respectively.

The directivity of the horn antenna is demonstrated qualitatively in Figure 4.3(b) which shows the spatial distribution of the real part of the electric field. The electric field propagates along the longitudinal plane (r, z) that contains the dipole source and the first 16 µm of the horn antenna. The real part of electric field shows the amplitude and phase properties of the optical waves that are channeled within the antenna. From the simulation results we can see that the propagating optical field within the flaring dielectric waveguide shows planar wavefronts and its amplitude distribution is a Gaussian-like shape. Such field property ensures highly directive propagations. Figure 4.3(c) and (d) show the free space emission diagrams of the two horn antennas as depicted above along two perpendicular longitudinal planes (y, z) and (x, z), respectively. The horn antenna is not connected to the fiber and it is considered to be placed in vacuum directly. The dipole represented in green in the figures is oriented along 0x. From Figures 4.3(c) and (d) we can see that the photons collected by the two horn antennas are radiated within cones semi-angles of 12° (red curve) and 13.5° (blue curve), with the antenna directivities of 160 and 230, respectively according to the equation 4.9. Like the properties of microwave horn antennas, the nano-optical horn antenna is able to efficiently convert dipolar emission into extremely directive radiations that can be efficiently outcoupled into free space or fiber modes.

The collection efficiency (horn tip quantum yield) (qtip) is defined as the ratio of the collected power confined within the tapered waveguide to the total radiation power, that is \[ q_{tip} = \frac{P_{out}(r = D/2)/P_r}. \] The collection efficiency (qtip) is shown in Figure 4.4(a), (b) as a function of the horn-mirror distance h ranging from 30 nm to 1270 nm for the two horn antennas parameters we depicted before. The wavelength range spans in the near-infrared...
Figure 4.3: (a) Power accumulated by two different geometries of optical horn antennas through the output π-plane as a function of the radial coordinate \( r \). (b) Spatial distribution of the real part of the electric field emitted by a dipole source coupled to an optical horn antenna. The field is displayed within the longitudinal plane \((r, z)\) that contains the dipole and the first 16\( \mu \)m of the flaring dielectric waveguide. (c,d) Emission diagrams \( P(\beta, \theta) \) when (c) \( \theta = 90^\circ \) \((y, z)-\)plane perpendicular to the dipole direction) and (d) \( \theta = 0^\circ \) \((x, z)-\)plane that contains the dipole direction), for the two different horn antenna geometries considered in (a) [169].

from 1000 nm to 2000 nm. According to the Figures 4.4 (a), (b), the maximum collection efficiency can be obtained when \( h \) is close to \( \lambda/4 \) for both the two horn antenna, which is in agreement with the empirical rules of microwave horn antenna design. The slight mismatch between the optimum \( h \) for the two horn antennas is due to the complex optical mechanism between the dipolar source and the antenna which is strongly dependent on the antenna parameters. From Figure 4.4 we can see that the collection efficiency is more than 78% over the spectral domain of 1000 – 2000 nm with the maximum value larger than 88% at 1000 nm wavelength.

The ability of the horn antenna to outcouple the collected photons into a SMF-28 fiber, which can be described by the coefficient \( q_{tip} C_m \), is shown in Figure 4.4(c) and (d). It
4.1. HORN ANTENNA FOR DIPOLE EMITTER COUPLING INTO OPTICAL FIBER

Figure 4.4: (a,b) Collection efficiency $q_{tip}$ as a function of $\lambda$ and the dipole-to-mirror distance $h$, for the two different horn antenna geometries considered in Figure 4.3(a). (c,d) Part of the radiated power from the dipole source $q_{tip}C_m$ that is collected and guided within the optical fiber by the two horn antenna with different geometries: coefficient $q_{tip}C_m$ as a function of $\lambda$ and $h$ [169].

represents the fraction of radiated power from the dipole source which is collected by the horn antenna and guided within the optical fiber. Figure 4.4 shows the ability as a function of $\lambda$ and $h$ for the two different horn antenna geometry. The result shows a maximum coupling efficiency between the dipole source and the optical fiber of more than 63% at the wavelength of 1450 nm. $q_{tip}C_m$ is still a little higher for the horn antenna with the curvature radius of 750 nm than that of 500 nm. For both configuration, the maximum photons are collected for horn-mirror distance $h$ close to $\lambda/4$.

Figure 4.5(a) shows the spectra of the total decay rate $\gamma_{tot}$ and the quantum yield $q$ of the emitter for the two horn antenna configurations. The emitter’s decay rate is not strongly enhanced and is rather a slowly increasing function of $h/\lambda$. One main drawback of the optical horn antenna is that it induces lower emission rates than free space when $h/\lambda$ becomes too small. From Figure 4.5(a) we can see that $\gamma_{tot} < 1$ when $\lambda > 1550\text{nm}$ for the horn antenna with $R = 500\text{ nm}$ and $h = 290\text{ nm}$. But the emission rate is not hindered over the spectral range when the horn antenna with curvature radius of 750 nm is employed. So the curvature radius should be chosen in a way that the optimum antenna-mirror distance is as large as possible to avoid weak emission configuration. The quantum yield is almost unaffected according to the simulation result shown in Figure (a).

Figure 4.5(b) and (c) show the spectra of parameter $T$, the probability that a photon emitted by the emitter is collected and guided within the optical fiber for the two horn antenna configurations: $R = 500\text{ nm}$ / $h = 290\text{ nm}$ and $R = 750\text{ nm}$/ $h = 390\text{ nm}$, respectively. For each configuration, the diameter $D$ of the output interface of the waveguide is varied from 9 $\mu$m to 12 $\mu$m by 1 $\mu$m step. $D$ has to be chosen to match the width of the fiber core mode. The influence of $D$ on the parameter $T$ is obvious from the simulation result. But the relationship between $D$ and $T$ is strongly dependent on the geometrical parameters of
Figure 4.5: (a) Spectra of total decay rate $\gamma_{\text{tot}}$ (dashed lines) and quantum yield $q$ (solid lines) of the structure for the two horn antenna geometries as shown in Figure 4.3. (b) spectrum of the overall photon transfer $T$ from the nanoemitter to the optical fiber, for $R = 500$ nm, $h = 290$ nm and four different values of parameter $D$ ranging from 9000 nm to 12000 nm. (c) spectrum of the overall photon transfer $T$ from the nanoemitter to the optical fiber, for $R = 750$ nm, $h = 390$ nm and four different values of parameter $D$ ranging from 9000 nm to 12000 nm [169].

So a trade-off has to be found between the geometrical parameters $R$, $L$ and $D$ of the dielectric waveguide to optimize the photon transmission in the optical fiber. From Figure (b) when $R = 500$ nm, $h = 290$ nm and $D = 10$ $\mu$m, $T$ keeps above 50% for all the wavelengths within the range of 1000 nm to 2000 nm and a maximum value of about 65% at wavelength of 1400 nm can be obtained. Figure (c) shows the situation when $R = 750$ nm, $h = 390$ nm and $D = 11$ $\mu$m, $T$ is larger than 50% within the wavelength range with a maximum value of about 70% at wavelength of 1200 nm. The horn antenna configuration with $R = 750$ nm and $D = 12$ $\mu$m is interesting for applications in telecommunications since it ensures a ratio of guided photons into the fiber larger than 60% over the spectral range of 1280 – 1750 nm with a maximum of 67% at wavelength of 1430 nm.
4.2. HORN FIBER TIP FABRICATION AND EXPERIMENTAL STUDY

4.2.1. HORN FIBER TIP FABRICATION

A horn fiber tip with the geometrical shape similar with the horn antenna theoretical model in previous section is fabricated. The fabrication of the horn fiber tip is based on a cleaved end facet of a SMF-28 optical fiber, which is a monomode fiber at the wavelength of 1550 nm with a numerical aperture of 0.14. The diameters of the fiber core and fiber cladding are 8.2 µm and 125 µm, respectively. The fabrication process is shown in Figure 4.6(a):

![Fabrication scheme of the dielectric horn](image1)

Figure 4.6: (a) Fabrication scheme of the dielectric horn, i.e. a fiber polymer microtip. (b) SEM image of a dielectric horn

Horn fiber tip is fabricated from photo-polymer, which can be polymerized by the laser light at the wavelength of 532 nm with the collaboration of Lovalite. The geometrical shape of the horn fiber tip is decided by the output fiber mode. Pure fundamental fiber mode in gaussian shape makes well-shaped horn tip. In our experiment, one droplet of photopolymer is put onto one cleaved end facet of the fiber and 532 nm laser is input from the other cleaved fiber end. Several small fiber loops should be made to select the fundamental fiber mode to ensure the perfect shape of the horn fiber tip. The output laser intensity to polymerize the material should be set at about 1.7 nW. The polymerization time should be carefully controlled since it decides the curvature radius of the horn fiber tip. Generally we set the time at about 10 s. After the illumination, the liquid polymer in illumination area is polymerized and the other liquid polymer out of the illumination area is removed using methanol. Then the horn fiber tip is obtained onto the fiber end facet with perfect alignment with fiber core. This horn fiber tip can effectively confine and guide light to the apex of the tip with a high-efficiency output. After the polymerization, a strong intensity laser about several mW should be used to illuminate the horn antenna to further polymerize it and also bleach the polymer material. The last step is to put the horn tip- integrated fibers into oven to cure them with high temperature of 180°C for 12 hours. The horn fiber tips with 500 nm curvature radius have been fabricated. Figure 4.6(b) is the SEM image of a horn fiber tip. The length of the horn tip is about 40 µm.

4.2.2. EXPERIMENTAL STUDY OF HORN TIP-INTEGRATED FIBER FOR IN-FIBER INFRARED FLUORESCENCE COUPLING

The horn tip-fiber system will be first applied to collect infrared QD fluorescence to compare the collection efficiency of the dielectric horn fiber system and conventional optical microscopy. In this study, the same colloidal PbS QDs (Evidot) as we introduced in chap-
ter 2 will be used. The experiment is based on an inversed optical microscopy shown in Figure 4.7.

In this experiment, the sample is prepared by spin coating the mixture of 5 µL original QDs dispersion (22.7 nmol/ml Evidot as introduced in chapter 2) and 90 µL PMMA A4 onto a microscopy glass with 5000 rps spin speed. Laser diode at 808 nm wavelength from Bluesky (VPS L0808 − 100X9B) is used to excite the QDs. The excitation light is collimated and passes through a short pass filter (FES850) and finally is focused onto the QDs sample by a 60×, 0.9 NA objective. A horn tip fiber is glued onto a tuning fork which is fixed onto SNOM system. The horn tip-to-sample distance can be kept at nanometer range using the SNOM. The horn tip fiber is placed at the center of the excitation beam and the fluorescence from the sample can be collected by the horn tip and conducted to photon counter (Aurea Technology SPD A with detection efficiency of 20%) through the fiber at last. To compare the collection efficiency of the horn tip and optical objective, confocal microscopy fluorescence detection in far-field is also recorded. A dichroic filter (Thorlabs, 850 nm) is used to separate detected fluorescence from the excitation laser. The fluorescence is focused into a single mode fiber (SMF-28) and conducted to the
4.2. HORN FIBER TIP FABRICATION AND EXPERIMENTAL STUDY

same photon counter, thus the signal level detected by the two methods can be compared directly. Note that a long pass filter (FEL1100) is fixed in a U-bench before the photon counter to remove the noise from the laser. The waveform generator and correlator in the experimental setup are used for measuring the decay curve of the QDs fluorescence, which will be introduced in the next part.

Based on the experimental setup, we first compare the collection signals from fiber and objective. In continuous excitation mode and with the excitation intensity of 3000 µW, 40 kcps photons are detected by the horn antenna fiber system while 4 kcps photons are collected by the confocal microscopy. The detection efficiency of the confocal microscopy is quite low, mainly because the excitation wavelength is very different from the emission wavelength and the confocal configuration is not proper for so large wavelength mismatch. With the fiber horn tip, 10 times more collection is realized.

RELATIONSHIP BETWEEN QD EMISSION AND EXCITATION INTENSITY

To study the relationship between the QDs fluorescence and the excitation intensity, we perform the experiment with horn tip collection. The relationship between the QDs fluorescence and the excitation intensity is shown in Figure 4.8.

![Figure 4.8: The relationship between the QDs fluorescence and the excitation intensity.](image)

The QDs fluorescence increases obviously with the increase of excitation intensity when the excitation intensity is smaller than 2000 µW. It is non saturation excitation. When the excitation intensity gets larger than 2000 µW, the fluorescence increases slowly until at last, it does not change any more with the increase of the excitation intensity, which is called saturation excitation.

Except the study of the collection efficiency comparison between horn tip and conventional microscopy, the relationship of the QDs fluorescence and excitation intensity, we also investigate the influence of excitation intensity, dielectric environment and QDs oxidation to the QDs fluorescence decay rate (excited state lifetime of QDs). For QDs fluorescence decay rate measurement, pulse mode excitation and pulse mode photon
counter detection are mandatory. In the experiments, the QDs are excited by pulse mode laser and the QDs photons are generally collected by fiber or objective and reaches the photon counter at last. Since it takes a long time (compare to the lifetime of the excited states) from the moment the laser pulse being generated to the moment photons reaching photon detector, the traveling time of electrical pulse, laser pulse and photons in optical path should be considered to measure the true lifetime of excited state, thus the pulse for triggering laser and the pulse for triggering the photon counter should be synchronized. Digital delay system is generally used to synchronize the two pulses as shown in Figure 4.9. The red path is for laser pulse transmission, QDs excitation and photon transmission and the blue path shows the gate pulse transmission to photon counter.

![Digital delay system of the experiment for QDs decay rate measurement.](image)

In our experiment for radiative decay curve measurement, both the laser and the photon counter work in pulse mode. A laser pulse with width of 5 ns is used to excite QDs and TTL (transistor-transistor logic) gate with gate width of 30 ns is used to trigger photon counter. During electrical pulse transmission in electrical cables and photons guiding in optical fibers, 1 m fiber corresponds to a time delay of 5 ns and 1 m electrical cable corresponds to 10 ns time delay. The digital delay between the two pulses can be evaluated based on these information. The pulse synchronization system in the experiment is included in the Figure 4.7. An arbitrary waveform generator (Agilent 33250A, 80 MHz function) is used for generating pulses to trigger laser and photon counter. Two electrical pulse outputs, one for the laser pulse generation and the other one for the photon counter gate pulse, are generated and synchronized by the generator. A correlator software based on computer is employed to record the arriving time of single photons since the moment that the QDs are excited. Thus the excited states lifetime of the QDs can be obtained.

**PMMA influence on the excited state lifetime**

Since nano emitters are generally 3D confined nanoscale structures with a large surface-volume ratio, the local environment of the emitters is very important concerning their optical properties, especially excited state lifetime. The influence of dielectric environment to the excited state lifetime of nano emitters has been investigated and reported by other group [172, 173, 174]. The theoretical and experimental results showed excited state lifetime change of emitters when they are placed inside a dielectric layer. Various formulas about the relationship between the excited state lifetime and the surrounding dielectric medium refractive index have been proposed. In Glauber’s research [173], the relationship between the radiative lifetime of emitters and the surrounding medium refrac-
The radiative lifetime of the emitters surrounded by external medium and the emitters exposed in air, respectively. $n$ is the refractive index of the surrounding medium.

To investigate the influence of the PMMA layer to the excited state lifetime of QDs, the excited state lifetime difference of the QDs droplet exposed to air and the QDs inside PMMA layer has been compared. Both the two lifetime measurements are based on the horn fiber tip collection and saturation mode excitation. The lifetime curves are shown in Figure 4.10. The experimental result shows that the excited state lifetime of the QDs embedded in PMMA is about 10 ns and the lifetime of the QDs exposed to air is about 23 ns. In our experiment, the refractive index of the surrounding medium (PMMA) of the QDs is about 1.5. According to the equation 4.10, we can get that the radiative lifetime of the QDs exposed in air is about 2.3 times of the QDs embedded in PMMA, which is consistent with our experimental results.

![Figure 4.10: The radiative lifetime of the QDs embedded inside PMMA layer (dark blue curve) and the QDs exposed in air (light blue curve).](image)

**Excitation intensity influence on the excited state lifetime in non-saturation excitation mode**

Based on the fiber horn tip collection, the excitation intensity influence to excited state lifetime change in non saturation excitation mode is investigated. The excited state lifetimes with three different excitation intensities are studied and in the three cases, the excitation intensities are $E_1$, $E_2$ and $E_3$ respectively and $E_1 > E_2 > E_3$. The radiative decay curves corresponding to the three cases are shown in Figure 4.11.

The radiative decay curves in Figure 4.11(a), (b) and (c) show the excited state lifetimes with excitation intensity of $E_1$, $E_2$ and $E_3$ respectively. The relationship between the three excitation intensities is: $E_1 > E_2 > E_3$. All the three excitation processes happened in non saturation excitation range. The experiment result shows that the lifetime in (a) is shorter than that in (b), and the lifetime in (c) is longer. So we can conclude that in non saturation
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Figure 4.11: Radiative decay curve when the sample is excited by three different excitation intensities. (a) with excitation intensity of $E_1$. (b) with excitation intensity of $E_2$. (c) with excitation intensity of $E_3$. $E_1 > E_2 > E_3$

excitation mode, the larger the excitation intensity is, the shorter the lifetime is, which is consistent with previous reports [175, 176].

The previous reports attribute the radiative lifetime change to biexciton generation with high intensity excitation. The radiative decay curves of exciton (X) and biexciton (XX) are shown in 4.12(a). From the result we can see that the radiative lifetime of biexciton is about half of the lifetime of exciton. The comparison between the radiative decay curves with lower intensity excitation (top figure of Figure 4.12) and higher intensity excitation (bottom figure of Figure 4.12) shows a higher biexcitation emission under higher intensity excitation [175]. Excitation intensity dependence of exciton X, biexciton (XX) and charged biexciton (XX*) amplitude has been investigated by Achermann’s group and the result is shown in Figure 4.12(b). The result shows that higher excitation intensity is more possible to generate biexciton in QDs. Woggon’s group also demonstrated that emission under high intensity excitation is characterized by the radiative processes connected with the biexciton system [177]. The combination of short lifetime of biexciton and higher possibility of biexciton generation under higher excitation intensity leads to the shortened radiative lifetime under higher excitation intensity, which is consistent with our experimental result.

OXIDATION INFLUENCE TO THE EXCITED STATE LIFETIME

The influence of oxidation of the QDs to the excited state lifetime change is investigated in this part. A sample with QDs exposed in air is used. The sample is simply prepared by putting a QDs droplet onto a cleaned microscopy cover glass. The excited state lifetimes
4.2. HORN FIBER TIP FABRICATION AND EXPERIMENTAL STUDY

Figure 4.12: (a) Radiative decay rate of an exciton (X) and biexciton (XX) for the same CdSe QD with lower excitation intensity (top) and higher excitation intensity (bottom) [175]. (b) Excitation intensity dependance of exciton X, biexciton (XX) and charged biexciton (XX*) amplitude compared to theoretical fitting curves (lines) [176].

of the sample at the first day and that after six days have been compared as shown in Figure 4.13. Figure 4.13(a) is the lifetime curve of QDs droplet at the first day. After six days being exposed to air, the lifetime curve changed to that as shown in Figure 4.13(b). The excited state lifetime is elongated obviously with the six days time exposed to air. As the comparison, lifetime of the QDs embedded in PMMA was also measured and after six days, the lifetime did not change. From the comparison between the two samples, we can get that the air environment changed the lifetime curve. Indeed, it’s the oxygen in air which leads to the lifetime change. The oxidation of the QDs would elongate the excited state lifetime. For the PMMA sample, the QDs are separated from air and could not be easily oxidized, so there is no obvious excited state lifetime change during the six days.

THE INFLUENCE OF GOLD MIRROR TO FLUORESCENCE DETECTION

This study corresponds to the nano-optical horn antenna concept exposed in the first theoretical part of this chapter. The influence of mirror to fluorescence detection has been analyzed in the simulation part. Simulation result shows that similar with the radiowave horn antenna, when the mirror is placed $\lambda f_{fluo}/4$ away from the dipolar emitter, maximum collection can be realized by the optical horn antenna. The constructive interference between the mirror reflected fluorescence and the unreflected fluorescence leads to the maximum collection. According to interference theory, periodic modulation of the detected signal with a period of $\lambda f_{fluo}/2$ is expected to observe. To verify this simulation, we perform an experiment. In the experiment, we directly put QDs droplet onto a cleaned microscopy cover glass. Then SNOM system is controlled to approach the horn tip to the QD droplet and attach some QDs onto the tip apex. Laser light at 808 nm (same laser source as we used in chapter 2 and 3) is guided to the horn tip by the optical fiber to excite the QDs. The fluorescence was also detected by the same horn tip. The same add-drop filter from Absys that we used in previous chapter is also used here. The filter is used eliminate
from the signal the excitation light. A mirror with Au film deposited is placed onto a holder which was controlled by a piezo stage system. The RHK control module introduced in chapter 2 is used to move the piezo stage in longitudinal direction, and thus it can control the horn tip-mirror distance. We approached the horn tip with QDs attached to the gold mirror until the distance is small enough in SNOM near field distance feedback control region (the distance is about several nanometer which can be recorded as "0") and we recorded the detected signal. Then the piezo stage is controlled to remove the mirror away from the QDs step by step with one step size of 20 nm and the detected photons are recorded for every step. The experimental results are plotted in the Figure 4.14.

The experimental results show a maximum collection at 0 nm QDs-to-mirror distance; periodic modulation of the signal is observed but with a period of about 140 nm, which is far from the expected period of $\lambda_{fllow}/2$ (that is about 750 nm due to the emission wavelength of 1500 nm). The discrepancy is mainly due to the parasitic residual fluorescence from the tip. When the dielectric tip material is illuminated by the light at 808 nm, infrared fluorescence from the material is detected by the photon counter, which strongly impacts on the collected fluorescence from the QDs. The parasitic residual fluorescence also limits the coupling efficiency of the horn tip from single dipole emitters. So the polymer horn tip is not applicable to all-fiber single photon source. The original purpose of single emit-
ter integrated horn antenna fiber system is not successfully demonstrated in experiments due to the residual fluorescence of the polymer horn tip. But we succeed in integrating the horn tip with scintillators for a new application of X-ray sensing.

4.3/ FIBER-INTEGRATED X-RAY SENSORS BASED ON IN-FIBER COLLECTION OF X-RAY EXCITED LUMINESCENCE BY NANO-OPTICAL HORN ANTENNA

This last study is a pioneering one aimed at developing a new technological approach in the engineering of X-ray sensors. The purpose is here to design and fabricate novel extremely compact and highly sensitive X-ray micro and nano-detectors at the end of single optical fibers, by exploiting the record emission directivity and in-fiber collection efficiency of nano-optical horn antennas. Main target outcome is the breakthrough design and implementation of novel architectures for X-ray real-time dosimetry. To this end, the horn tip fiber bench is integrated with X-ray excited luminescent materials, instead of fluorescent quantum dots, to detect the X-ray excited luminescence.

The main idea is to engineer the nano-optical horn antenna concept for efficiently collecting and transferring X-ray Excited Photoluminescence (XEP) from nanoscintillators towards optical fibers, in order to achieve high performance X-ray detectors available in compact and flexible architectures free from bulky optics and compatible with endoscopy. With this upstream study and development, nano-optics will make a first key contribution to the development of X-ray sensing protocols and architectures. In that context, NOAs would open the way towards radically new concepts allowing for “on-fiber” X-ray micro and nano sensors for in situ and in vivo scientific, medical and industrial metrology and characterization. By leveraging the versatility and ubiquity of fiber-optics technology, this
may constitute a key step towards the widespread use of endoscopic techniques in X-ray digital radiography and real-time dosimetry.

In this section, we will first introduce the state-of-the-art of fiber X-ray sensing. Then, we experimentally realize integration of luminescent particles (scintillators) onto the fiber tip. Finally, this new concept of tiny fiber integrated X-ray sensor is demonstrated with the characterization of a the profile of a focused X-ray beam at 8-10 keV energies.

4.3.1 State-of-the-art in the domain of fiber X-ray sensing

X-rays are nowadays essential in the characterization and metrology of materials, objects and living species as well as in cancer therapy. The industrial development of X-ray detectors is however still hindered by the difficulty to achieve efficient “X-photon”-to-electron conversion in electronic devices. Indirect detection technique, which combines luminescent materials to silicon-based optical detectors [178], has demonstrated performances in terms of image contrast and signal dynamics, and is now widely developed by many companies. Problem remains however that the resulting sensors and cameras suffer from modest spatial resolution (about 10 microns) and from problems of cross-talk between neighbouring pixels. Best resolution up to 1.6 microns is achieved with Rigaku cameras which incorporate bulky magnifying objectives [14]. On the other hand, X-ray sensors and cameras are often too bulky to allow easy handling, rapid implementation into systems and endoscopic applications.

Plastic scintillator-based fibered dosimeters have been widely studied for use and applications in radiation therapy measurements [179], in architectures compatible with endoscopy [180], radiation monitor [181] and so on. These devices consist of millimeter scintillator capsules embedded in a module connected to a multimode fiber. They allow for fiber-assisted X-ray excited photoluminescence (XEP) transfer from the scintillators to a remote optical detector. Previous work has shown the ability of fiber-optic dosimeters that feature 5 mm-long plastic scintillators to measure small radiation fields down to 1 cm diameter. Fiber-optic sensors assembled in 2D arrays have been demonstrated to measure radiation field sizes down to 1.5 cm × 5 cm with a resolution of 5 mm [182]. Researchers have used a plastic scintillator-based fiber-optic dosimeter in translational movement with a stepper motor to acquire the 1D lateral dose profile of a 10 cm × 10 cm electron beam at a resolution of 3.9 mm [183]. However, the overall dimensions and resolution of these detectors are typically larger than 1 mm. These systems are thus not compatible with high resolution digital radiography and real-time dosimetry, as well as X-ray camera engineering due to cross-talk effect.

The development of a microscale resolution endoscopic dosimeter has been published in 2015 for X-ray micro-beam radiation therapy [184]. However, the X-ray sensor in play, taking the form of a 11 micron thick pellet covering the 600-micron diameter core of a multimode fiber, leads to microscale resolution (10 microns) for a single incidence angle perpendicular to the fiber axis. Deviating the incidence angle by only 8 results in a 10-fold spatial resolution decrease. Moreover, the overall bare probe (i.e. not encapsulated) is still wide on a few hundreds of millimeter range (close to millimeter range). The use of a large core optical fiber is here imposed by the very low coupling efficiency between optical fibers and dipolar luminescent sources. Moreover, this technique has not been applied to conventional broad beam radiation therapy yet, and is definitely not suitable to design X-ray endoscopic cameras due to resolution and cross-talk limitations.
To date, as noted in the introduction chapter, in-fiber XEP transmission with monomode fiber has been reported by CINAM partner [55, 185]. CINAM performed luminescence mapping, simultaneously with sample topography, in a collection-mode SNOM experiment involving metal coated fiber tips whose apex is opened with a single 50 nm wide aperture. Luminescence mappings with 50 nm spatial resolution have been demonstrated with photomultiplier detection and a low power laboratory X-ray tube (800 A at 35 kV, Rh target X-ray source). These results are important for this study because they show that the intrinsic yield of scintillators and the efficiency of the existing optical detectors are high enough to detect XEP signals from nanoscale luminescent volumes, limited here by the size of the low-throughput aperture. The implementation of fiber nano-optical horn antenna should strongly increase the luminescence detection performances of the overall system.

### 4.3.2 Fiber X-ray sensor fabrication: optical horn tip + scintillators

The fabrication of fiber-integrated X-ray nano-detectors is cascaded in three steps, namely (1) the fabrication of the dielectric horn at the end of a cleaved fiber (see section 4.2.1), (2) the attachment of single micron-size luminescent particle at the apex of a fiber polymer horn, and (3) metal-coating with possible preliminary dielectric spacer coating. Such a process leads to X-ray excited luminescent nano-optical horn structures.

Particle attachment is realized by mechanical contact between dielectric horn apex and single particle deposited onto flat substrate, following a process already demonstrated for the grafting of fluorescent elements at the apex of SNOM tips [186]. We adapted the process as shown in Figure 4.15.

![Figure 4.15](image)

Figure 4.15: (a) Experimental scheme for grafting luminescent particles onto fiber dielectric horns. (b) Close view of the part shown in the dashed circle in (a).

The experimental setup is based on an inversed optical microscope. The sample is prepared by spreading particles onto an area of a substrate. Another area of the substrate is covered by UV glue layer (Norland Optical UV glue 63), which can be polymerized by UV illumination. The sample is fixed to a prism as shown in Figure 4.15. The prism is mounted onto a holder which is fixed onto a piezo stage. The RHK control module is employed to control the piezo stage in 3D with very high resolution (5 nm movement resolution in x and y direction with a total travel range of $10 \times 10 \, \mu m^2$; 5 nm movement resolution in z direction with a total travel range of 5 µm). The fiber horn tip is mounted.
onto 3D translation system (MAX350D/M translation system from Thorlab) with 50 nm movement resolution and 300 μm travel range for the coarse adjustment to approach the horn tip to the sample. An objective of long focal length 125×, 0.8NA (LEITZ PLAN L 125 × 0.80) is used to image the sample together with the horn tip onto a visible camera in real time. During the experiment, we approach the fiber to sample by coarsely adjusting the 3D translation system. The distance information between the fiber and the sample can be got from the camera. Then the RHK control module is used to move the sample to contact with the horn tip with fine resolution. Very careful operation for the particle attachment is made due to the weakness of the horn tip. The fiber is first contacted to the UV glue to attach a little glue onto the tip apex. After we move the fiber to the particle area to attach the particles. Since the glue is viscous, with the glue on the tip, it is easy to attach the particles experimentally. Finally, we solidate the glue using a UV light.

Such a process has been first tested in the attachment of SiO₂ beads with size of 1 μm, 500 nm and 250 nm. The SEM images are shown in Figure 4.16. The two larger particles have been attached onto metal coated polymer microtips (with some of them fabricated to receive BNA at their apex (see Figure 4.16(c) and (d))). Such tips avoid charge accumulation at the tip apex which can occur for dielectric bare horns. 1 μm diameter SiO₂ particle has been attached as shown in Figure 4.16(a). The attachment of the SiO₂ particles with diameter of 500 nm have been shown in Figures 4.16(b), (c) and (d). The small dark areas in the Figures are the UV glue areas. At this particle diameter, the trapped bead starts to be decentered with respect to the fiber tip axis. This phenomenon is due to the sample tilt with respect to the microtip axis and to rounded tip apexes larger that the bead diameter. Centered particles can thus be expected for particle diameter of the order of the apex size. Figures 4.16(e) and (f) show enhanced particle decentering, which is consistent with our interpretation. These SEM images also show little tip charging during SEM imaging, the particle appears as a semi sphere and only slight roughness is observable onto the tip. This is because bare dielectric horns have been used here and structure charging can occur in that case. This could be reduced by inserting conducted silver paste in-between the fiber and holder (the holder used to support the fibers in SEM) during SEM imaging process.

Figure 4.17 shows tiny clusters of ZnO luminescent particles attached at the end of (a) metal-coated and (b) bare fiber dielectric horns. The aggregates are composed of particles with diameters smaller that of about 200 nm. Note that it is a little more difficult to attach a single ZnO cluster than a single SiO₂ particle due to the irregularity of the clusters shape. No charge accumulation occurs at the bare dielectric horn apex as the image contrast and resolution are optimum. However, horn apex lost its initial well defined shape (as if the polymer melted). This is due to SEM imaging process which sometime affects the polymer. Apex shape modification gets stronger and stronger during multiple image acquisition process. Therefore, scintillator grafted dielectric horns used for sensing are not images with SEM, only at optical microscope.

### 4.3.3 Experimental demonstration of X-ray sensing

Metal layer is then deposited by evaporation onto the overall structure to finalize the fiber nanoscale sensor. The type of metal as well as particular fabrication processes are not detailed due to possible patents opportunities related to this work. The resulting fiber "nano-optical horn tip/scintillator" platform is shown in Figure 4.18.(a). It is ultra-compact,
3. FIBER-INTEGRATED X-RAY SENSORS BASED ON IN-FIBER COLLECTION OF X-RAY EXCITED

Figure 4.16: SEM images of the SiO$_2$ particles on horn tips. (a) SEM image of the horn tip with 1 µm diameter SiO$_2$ particle attached. (b), (c) and (d) are SEM images of the horn tips with 500 nm diameter SiO$_2$ particles attached. (e) and (f) are SEM images of the horn tip with SiO$_2$ diameter of about 250 nm. The scale bar in the four images is 2 µm.

Figure 4.17: SEM images of the ZnO particle aggregates onto two dielectric horn apex. The scale bar is 500 nm.

fiber-integrated and plug-and-play.

Figure 4.18(a) shows the experimental scheme for fiber X-ray sensor demonstration. Experiments have been realized in collaboration with CINAM which developed X-ray imaging bench. A Rh X-ray tube source emits X-rays at energy range 5-30 keV. The high energy radiations are focused with a polycapillary lens. The experiment is performed with the
Figure 4.18: (a) Experimental scheme for X-ray sensing. (b) Time trace of the detection signal with the fiber sensor placed within an X-ray focused beam, at focus, and with X-ray source on and off. (c) Experimental cross-section plot of the X-ray focal spot. (d) Calibration curve of the dosimeter (detected luminescence vs incident X-ray flux). Minimum detectable X-ray flux is estimated to 100 photons/s/µm². (e) Luminescence spectrum of the dosimeter (i.e., of the scintillator cluster embedded into the fiber nano-optical horn antenna.

X-ray source current lower than 800 µA. The fiber X-ray sensor is positioned near focus plane, centered with respect to the system optical axis, and the free fiber output is connected to a photon counter from Aurea Technology. The beam flux has been measured to about $2 \times 10^9$ photons/s and the FWHM of the beam profile near focus is at 25 microns (manufacturer’s data). Figure 4.18(b) shows the acquisition time trace with the X-ray source on and off. The signal-to-noise ratio is here very high with signal level during sensor irradiation of the order of 120 kcounts/sec, which is much higher than the noise level of a few tens of counts/s. Then the fiber sensor is displaced transversely with respect to the beam propagation axis to plot beam profile about the focal spot (Figure 4.18(c)). The FWHM measured from experimental plot is of 26 microns. Figure 4.18(d) displays the response of the dosimeter on fiber as function of the impinging X-ray flux. We clearly show a linear response of the system with respect to the X-ray photon dose. This demonstrated the ability of our system to work as a local dosimeter of X-ray radiations. To ensure that the optical signal detection is due to the scintillators embedded into the
structure and not induced by the polymer of the fiber itself, we measured the spectrum of the collected light (Figure 4.18(e)). The experimental curve is obtained by connecting the fiber output to an optical spectrum analyzer (Princeton SP2300) which consists of a grating coupled to a photomultiplier. We see that the X-ray excited luminescence collected through the fiber is centered to 530 nm. This is the spectral signature of the scintillator coupled to the nano-optical horn antenna, which validates our X-ray nano-optical sensor concept. Indeed the emission spectrum of the scintillators is predicted to be centered 520 nm. The slight wavelength shift between what we measure and the could be either due to the spectrum analyzer or to a redshift of the emission due to the scintillator embodiment into the nano-optical horn antenna (contact with the polymer and metal layer).

Note that, so far, profiling X-ray focused beams stays very challenging with conventional techniques based on pinhole coupled with a Geiger counter [187]. The resulting bench is composed of various components that have to be aligned one from each others and also with respect to the invisible ionizing beam. Our ultra-compact and plug-and-play fiber X-ray sensor highly simplifies the measurements. Since the detection bench is auto-aligned, beam profiling becomes very straightforward and new complete information onto beam shape becomes possible. Our fiber-integrated nano-optical horn antenna based system thus offers very promising scientific but also medical and industrial perspectives in X-ray beam characterization. Further applications are also targeted in digital scanning radiography.

4.4 CONCLUSION

In this chapter, we presented and developed the concept of nano optical horn antenna to couple the photons from a single dipolar emitter efficiently into an optical fiber. Theoretical simulation results a coupling efficiency as high as 85% for the emitter with the fluorescence wavelength of 1.45 µm. A horn tip similar with the horn antenna model in the simulation was fabricated onto a single mode fiber end facet. But the fluorescence collection from a single QD was not successfully realized due to parasitic residual fluorescence from the tip material. So we developed a new application of the horn tip as X-ray sensor. In this application, the horn antenna was integrated with scintillator to collect the X-ray excited luminescence efficiently and used as the first ultra-compact fiber-integrated X-ray real-time dosimeter. This is the first use of nano optical tips to control emission of X-ray excited luminescence to couple high efficiency luminescence into optical fibers, with the promising applications in radiation therapy measurement, in architectures compatible with endoscopy, radiation monitor and so on.
In this thesis, we mainly focused on developing flexible, compact, plug-and-play optical fiber systems for luminescence detection (QD telecommunication fluorescence and X-ray excited luminescence) and imaging down to single emitters. Different kinds of nano antennas and axicons have been proposed as the interfaces of luminescence materials and optical fibers to transfer luminescence into optical fiber efficiently.

At first, a single mode optical fiber-based AXIGRIN has been proposed for far-field in-fiber fluorescence coupling, which has been demonstrated with the use of non-diffracting Bessel beams, thanks to an ultra-compact and self-aligned axicon. Such beams are shown to provide the first resolved infrared fluorescence imaging of PbS quantum dots (808 nm excitation wavelength and 1502 nm emission wavelength), where spherical optics fails due to the problems of chromatism.

Then, in-fiber near-field coupling is achieved by means of the concept of double resonance BNA, with demonstration of nanometer resolution all-fiber near-field imaging of single PbS (infrared) quantum dots. The two resonance wavelengths of the BNA were set at the QDs excitation wavelength and QDs fluorescence wavelength, thus both the excitation light and fluorescence of the QDs can be enhanced with the BNA. Such fiber-integrated near-field microscopy bench is ultra-compact since it is fully fiber integrated and it avoids the use of bulky optics: fluorescence excitation and collection are realized locally down to the nanometer scale through the same fiber. Experimental result demonstrated single PbS QDs imaging with spatial resolution as high as \( \lambda_{em}/20 \).

At last, the concept of fiber nano-optical horn antenna was proposed and investigated for in-fiber luminescence out-coupling. First investigation concerned the generation of fiber-integrated single photon sources. The concept was also demonstrated in the coupling of X-ray excited luminescence into single-mode fibers, with the purpose of generating the first generation of fiber X-ray sensors and dosimeters. We explore here for the first time the concept of nano-optical antenna to control X-ray excited luminescence (here directivity). Generally, nano-optical antennas are "limited" to the control of fluorescent emitter, not yet X-ray sensitive scintillators.

In one word, three optical fiber systems have been developed for luminescence detection. The concerned applications of these optical systems are fluorescence microscopy, sensing and fibered single photon detection.
12. Huang, Xiaoyong and Han, Sanyang and Huang, Wei and Liu, Xiaogang, “Enhancing solar cell efficiency: the search for luminescent materials as spectral converters,” Chemical Society Reviews, 42 number 1, 173–201 (2013)
14 "http://www.rigaku.com/en"


19 Pan, Caofeng and Dong, Lin and Zhu, Guang and Niu, Simiao and Yu, Ruomeng and Yang, Qing and Liu, Ying and Wang, Zhong Lin, “High-resolution electroluminescent imaging of pressure distribution using a piezoelectric nanowire LED array,” Nature Photonics, 7, number 9, 752-758 (2013)


34. Jeurissen, Ben and Leemans, Alexander and Tournier, Jacques-Donald and Jones, Derek K and Sijbers, Jan, “Investigating the prevalence of complex fiber configurations in white matter tissue with diffusion magnetic resonance imaging,” Human brain mapping 34, number 11, 2747–2766 (2013)


41 Cuche, Aurelien and Masenelli, B and Ledoux, G and Amans, D and Dujardin, C and Sonnefraud, Yannick and Melinon, P and Huant, Serge, “Fluorescent oxide nanoparticles adapted to active tips for near-field optics,” Nanotechnology 20, number 1, 015603 (2008)


46 Hennessy, T and Busch, Th, “Detecting atoms trapped in an optical lattice using a tapered optical nanofiber,” Optics express, 22 number 26, 32509–32519 (2014)

47 Fujiwara, Masazumi and Toubaru, Kiyota and Noda, Tetsuya and Zhao, Hong-Quan and Takeuchi, Shigeki, “Highly efficient coupling of photons from nanoemitters into single-mode optical fibers”, Nano letters 11, 4362-4365 (2011)

48 Schro¨der, Tim and Fujiwara, Masazumi and Noda, Tetsuya and Zhao, Hong-Quan and Benson, Oliver and Takeuchi, Shigeki, “A nanodiamond-tapered fiber system with high single-mode coupling efficiency,” Optics express 20, number 10, 10490– 10497 (2012)

49 Davanc, o, Marcelo and Srinivasan, Kartik, “Fiber-coupled semiconductor waveguides as an efficient optical interface to a single quantum dipole,” Optics letters 34 number 16, 2542-2544 (2009)


58 Schietinger, Stefan and Barth, Michael and Aichele, Thomas and Benson, Oliver, “Plasmon-enhanced single photon emission from a nanoassembled metal-diamond hybrid structure at room temperature,” Nano letters 9, 1694-1698 (2009)


70 Lu, Guowei and Li, Wenqiang and Zhang, Tianyue and Yue, Song and Liu, Jie and Hou, Lei and Li, Zhi and Gong, Qihuang, “Plasmonic-enhanced molecular fluorescence within isolated bowtie nano-apertures,” Acs Nano, 6, 1438-1448 (2012)
71 Mivelle, Mathieu and Viktorovitch, Pierre and Baida, Fadi I and El Eter, Ali and Xie, Zhihua and Vo, Than-Phong and Atie, Elie and Burr, Geoffrey W and Nedeljkovic, Dusan and Rauch, Jean-Yves and others, “Light funneling from a photonic crystal laser cavity to a nano-antenna: overcoming the diffraction limit in optical energy transfer down to the nanoscale,” Optics express 22, number 12, 15075–15087 (2014)
73 El Eter, Ali and Hameed, Nyha M and Baida, Fadi I and Salut, Roland and Filiatre, Claudine and Nedeljkovic, Dusan and Atie, Elie and Bole, Samuel and Grosjean, Thierry, “Fiber-integrated optical nano-tweezer based on a bowtie-aperture nano-antenna at the apex of a SNOM tip,” Optics express 22, number 8, 10072–10080 (2014)
77 Molenda, D and des Francs, G Colas and Fischer, UC and Rau, N and Naber, A, “High-resolution mapping of the optical near-field components at a triangular nano-aperture,” Optics express, 13, number 26, 10688–10696 (2005)


104 Grosjean, T and Sabac, A and Courjon, D, “A versatile and stable device allowing the efficient generation of beams with radial, azimuthal or hybrid polarizations,” Optics communications 252, number 1, 12–21 (2005)


147 Gerelli, Emmanuel, “Nanopinces Optiques a base de Modes de Bloch lents en Cavite,” INSA, Lyon (2012)


151 Mivelle, Mathieu, “Etude et de`veloppement de nano-antennes fibre´es pour la microscopie en champ proche optique et la nano-photonique,” (2001)
152 Guo, Hongcang and Meyrath, Todd P. and Zentgraf, Thomas and Liu, Na and Fu, Liwei and Schweizer, Heinz and Giessen, Harald, "Optical resonances of bowtie slot antennas and their geometry and material dependence," Optics Express 16, number 11, 7756–7766 (2008)


156 Wenger, Je rô me and Ge rard, Davy and Dintinger, Jose´ and Mahboub, Oussama and Bonod, Nicolas and Popov, Evgeny and Ebbesen, Thomas W and Rigneault, Herve’, "Emission and excitation contributions to enhanced single molecule fluorescence by gold nanometric apertures" Optics express 16, number 5, 3008–3020 (2008)

157 Girard, Christian and Martin, Olivier JF and Le ve que, Gae´ tan and des Francs, Ge rard Colas and Dereux, Alain, "Generalized bloch equations for optical interactions in confined geometries," Chemical physics letters, 404, number 1, 44-48 (2005)


159 Jaksic’, Vojkan and Pillet, C-A, "On a model for quantum friction, II. Fermi’s golden rule and dynamics at positive temperature," Communications in Mathematical Physics, 176, number 3, 619-644 (1996)


167 Silver, Samuel, “Microwave antenna theory and design,” number 19 (1949)


178 Granfors, Paul R and Aufrichtig, Richard, “Performance of a 41 × 41-cm2 amorphous silicon flat panel x-ray detector for radiographic imaging applications,” Medical physics, 27, number 6, 1324-1331 (2000)


185 Fauquet, Carole and Dehlinger, Mae”l and Jandard, Franck and Ferrero, Sylvain and Pailharey, Daniel and Larcheri, Sylvia and Graziola, Roberto and Purans, Juris and Bjeoumikhov, Aniouar and Erko, Alexei and others, “Combining scanning probe microscopy and X-ray spectroscopy,” Nanoscale research letters, 6, number 1, 1-6 (2011)


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Z. Xie, T. Grosjean, “Horn antenna development for all-fiber X-ray excited luminescence detection”, in preparation
Abstract:

My thesis is devoted to develop ultra-compact, plug-and-play and low-cost single-mode optical fiber systems for in-fiber luminescence collection. First, a new fiber self-aligned axicon is proposed to provide the first resolved infrared fluorescence imaging of PbS quantum dots in far field. Then, all-fiber near-field imaging of single PbS quantum dots is achieved by double resonance bowtie nano-aperture antenna (BNA) with nanometer resolution. Finally, the concept of fiber nano-optical horn antenna is proposed for in-fiber X-ray excited luminescence out-coupling, with the purpose of generating the first generation of fiber X-ray sensors and dosimeters.

Keywords: nanoantenna, optical fiber, SNOM (Scanning near-field optical microscopy), horn antenna, BNA antenna, AXIGRIN lens, nanoemitter, quantum dot

Résumé :

Ma thèse concerne le développement de systèmes ultracompactes auto-alignés et à faible coût intégrés sur fibre optique monomode pour la collection intégrée de la luminescence locale. Dans un premier temps, un axicon fibre auto-aligné (AXIGRIN) est proposé permettant de fournir la première imagerie résolue ultracompacts fibre de quantum dots PbS infrarouges. Ensuite, la première nano-imagerie (système entièrement fibre) de quantum dots PbS uniques est réalisée à l'aide d'une nano-antenne à ouverture bowtie intégrée sur pointe fibre. Enfin, le concept d'« antenne cornet » nano-optique est proposé pour le couplage direct et efficace de la luminescence excitée par rayons X à une fibre optique, dans le but de générer les premiers capteurs et dosimètres à fibres à rayons X.

Mots-clés : nano-antenne, fibre optique, SNOM (Scanning near-field optical microscopy), antenne cornet, nano-antenne à ouverture bowtie, lentille AXIGRIN, quantum dot infrarouge PbS, émetteur quantique