MASTER

Directional light emission from nanowire structures

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Abstract

A central topic of nanophotonics is to control the light emission by exploiting the interaction between different emission sources and nanostructures. As typical one-dimensional nanostructures, nanowires have proved to be a promising candidate to shape the property of light emission, either from itself or from other dedicated sources in its vicinity. In this thesis we will investigate the emission directivity of two different nanowire structures, namely vertical nanowire dimers and horizontal single nanowires. We will demonstrate that the emission directivity from a nanowire can be tailored by designing a multi-nanowire ensemble or by confining the source position inside the nanowire. We believe that the results of this thesis are meaningful for the purpose of designing nanowire-based light sources such as nanowire LEDs and single photon sources.
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Chapter 1

Introduction

1.1 Directional light emission at the nanoscale

For multiple purposes, from simple daily illumination to profound photon sources for quantum computing, people have been endeavouring for centuries to study the fundamental mechanisms, quest for novel materials and engineer the patterns of light emission. In particular, plenty of specific applications demand for a directional light source. Traditionally, bulky optical components such as lenses and mirrors are utilised to control the direction of light. Nowadays, thanks to the rapid development of nanotechnology during the past a few decades, scientists are able to literally design, fabricate and characterise various nanostructures. Naturally, more interests are being paid to probe the way to achieve directional light emission in the subwavelength region, where a strong interaction between light and matter is expected.

Many novel devices could potentially benefit from the control over the directivity of light emission at nanoscale. For example, a directional LED could be highly valued in some specific situations (such as automobile light systems) while unidirectional light emission could significantly improve the efficiency of single photon sources. Therefore, the attempt of harnessing light emission with variable nanostructures has unsurprisingly become a research hot spot in the physics and optics communities all over world. In this thesis, we investigate the directional light emission from a particular nanostructure: the nanowire, which has draw much attention recently due to its competence of controlling the light emission.
1.2 Emitters in a cavity

In general, the light emission from a nanoscaled structure can be considered as emitters located in a 'cavity'. Here the cavity is an umbrella term, in which any metallic or dielectric micro-nano structures that are capable of effectively confining the electromagnetic fields can be included. The emission property depends dramatically on the interaction between the emitters and the cavity. Essentially, a certain emitter, which by nature is a sophisticated quantum mechanical system, can be abstracted as a two-energy-level system as schematically shown in figure 1.1. When the system is excited to the high energy state somehow and subsequently relaxes to the lower energy state, photons with energy equal to the energy difference between the two levels \(\hbar \omega\) can be emitted (assuming the transition is radiative). Fermi’s golden rule prescribes the decay rate of the system \(\gamma\), which is defined as the probability of transition to the ground state per unit time\([1]\):

\[
\gamma = \frac{2\pi}{\hbar^2} \int d\omega_f |\langle i|\hat{H}|j\rangle|^2 \delta(\omega_f - \omega_i)
\]

where \(\hat{H}\) is the perturbatory interaction Hamiltonian and usually approximated as electric dipole interaction \(\hat{H}_{dipole} = -\vec{\mu}_{ij} \cdot \vec{E}\) with \(\vec{\mu}_{ij} = e \langle i|\vec{r}|f\rangle\) being the dipole transition moment.

![Figure 1.1: Light emission from two-energy-level system](image.png)

In the scenario of spontaneous emission, an external electromagnetic field is absent and thus classically the transition could not occur due to the lack of necessary perturbation. However, the difficulty could be resolved once the electromagnetic field is also quantised. One need resort to quantum electrodynamics for the rigorous treatment, which is beyond the scope of this thesis. But intuitively and classically, it can be postulated that there still exists an vacuum field energy \(E_{vac}\) when no external electromagnetic
field is practically applied at all. Then the Fermi’s golden rule could be qualitatively valid for the spontaneous dipole emission. In this case, it could be written as

$$\gamma = \frac{2\pi}{h^2} | \langle i | \vec{\mu}_{ij} : \vec{E}_{\text{vac}} | j \rangle |^2 \rho(\omega)$$  \hspace{1cm} (1.2)$$

where $\rho(\omega)$ is the final local photonic density of states.

It is justified from equation 1.2 that one can differentiate the contributions to the spontaneous emission (characterised by decay rate $\lambda$) between the emitter and the ‘cavity’ environment, assuming that the interaction is in the weak coupling regime. The matrix term describes the dipole emission from the emitter itself while the photonic density of states is dictated by the environment where the emitter resides. The significant inspiration from this differentiation is that the light emission can be manipulated by designing an appropriate surrounding for the emitter.

Now that the freedom of modifying the light emission through the photonic environment is clear, what kinds of modification could be achieved? The most evident one is the rate of the emission, which determines the intensity of light emitted. From equation 1.2, it can be judged that if the photonic density of states reaches its peak value near the emission spectrum of the emitter, e.g. the emitter is tuned in frequency with a certain resonance, the emission will be considerably enhanced. This enhancement is called Purcell Effect, which is first analysed by Purcell based on the atom emission inside a traditional resonant cavity. Since then, various cavity configurations and resonant effects have been exploited to enhance the spontaneous emission, such as photonic crystals[3], plasmonic structures[4] and nanowire waveguides[5], to name but a few.

The second character of emission which can be modified due to a certain environment is its directivity, on which the research in this thesis is focused. Commonly, the discussion on the emission directivity is based on the far-field radiation (emission) pattern, which is defined as radiated power per unit solid angle. As introduced above, the emission from a emitter in most cases is described quantum mechanically by a electric dipole transition moment, which can be simply modelled as an oscillating electric dipole in the frame of classical electrodynamics. In the most featureless environment-vacuum, the far-field dipole emission pattern is given by[6]

$$\frac{dP}{d\Omega} = \sqrt{\frac{\mu_0}{\epsilon_0}} \frac{c^2}{32\pi^2} k^4 |\vec{p}|^2 \sin^2 \theta$$  \hspace{1cm} (1.3)$$
Obviously, the far field emission from a single dipole with defined direction is endowed with an angular dependence of $\sin^2 \theta$, where $\theta$ is the angle between the dipole moment vector $\vec{p}$ and the observation position $\vec{r}$. However, when we consider a emitter spontaneously radiates in vacuum, the direction of the dipole model is completely random. That means that the pattern of overall emission should be generated statistically from three orthogonal dipoles with identical weight and thus becomes omnidirectional (isotropic). In contrast, when being in a certain photonic environment, the directionality of the emitted light could be changed due to favourable coupling of emission to certain photonic modes. In this case directivity and the enhancement of the emission cannot be completely separated since the directivity is introduced naturally through the enhancement or inhibition of emission to different modes, which themselves show different farfield directivity. However, apart from enhanced coupling to particular modes, light emission with different farfield directivity can also be achieved through secondary scattering in the nearfield. Various structures utilizing this approach have been proposed, with the so-called optical Yagi-Uda antenna being a typical example\[7\]. While the scatterers used in the previous studies are primarily metallic particles\[8\][9], we will demonstrate in the thesis that semiconductor nanowires could also be used as scatterers to direct light emission.

It should be noted that for practical applications simply taking the isotropic emitter as a reference to discuss the emission directivity of other emitters is scarcely appropriate. Because any existence of necessary structures, as simple as a substrate for laying fluorescent materials, will modify the directivity of emission. Consequently, the reference and thus the figure of merit concerning the emission directivity should be considered specifically for certain practical devices.

### 1.3 Nanowire photonics

As discussed above, the underlying physics allows us to manipulate photons in the subwavelength regime, where the research in the field of nanophotonics is centred. Among a variety of micro-nano structures and materials being employed, the semiconductor nanowires emerged at the end of 20th century have shown great potential for this purpose. A couple of peculiarities make nanowires prominent for nanophotonics. First of all, they feature a high-aspect ratio. By definition, nanowires form a class of semiconductor nanostructure with cross-sectional dimensions varying from
several to hundreds of nanometre while length spanning from hundreds of nanometres to millimetres. This high aspect-ratio decides that nanowires can be used to bridge the nanoscopic and microscopic world, which is critical for the integration of nanoscale blocks into electrical or optoelectronic devices[10]. Furthermore, the optical properties of nanowire can be easily tuned either by changing their geometric dimensions or by engineering their atomic compositions. Nowadays, as a research field dedicated to study the distinct optical properties of nanowires and explore the possibility of their applications, nanowire photonics has been categorised as an unique branch of nanophotonics.

Since the first III-V semiconductor nanowire was synthesized at Hitachi[11] in 1990s, a couple of methods have been successfully implemented to grow nanowires with a broad range of compositions. Thus far the most widely used one remains the ‘vapour-liquid-solid’(VLS) technique, which uses catalytic liquid metal particles to initiate and orient the growth, as schematically described in figure 1.2(a). Recent understanding and development on VLS method has make it possible to precisely control the length, diameter, growth direction and morphology of the nanowires during growth. More dramatically, the atomic composition of the nanowire is highly tunable with VLS method as well, which makes it possible to grow doped nanowires or nanowire heterostructures and fabricate diverse nanowire-based devices. Apart from individual nanowires, highly ordered nanowire arrays with patterns on demand could also be grown by precisely controlling the position of seeding particle with lithography methods[12]. Nevertheless, the use of metallic particles in the VLS growth process leaves the synthesized nanowires inevitably contaminated, which is a detrimental problem for the efficiency of some optoelectronic devices[13]. To avoid this problem, several alternative catalyst-free methods are developed. A well-known one is selective-area metal-organic vapour phase epitaxy (SA-MOVPE), initially attempted at Hokkaido University[14]. As is illustrated in figure 1.2(b), SA-MOVPE essentially corresponds to an epitaxial layer growth through openings in a mask film deposited on the substrate. The openings are prepared ahead of growth by e-beam or nanoimprint lithography and chemical etching, which means their patterns could be freely designed and precisely controlled according to one’s demand.

The enormous success in growing high-quality nanowires has paved the path for their applications. And indeed a large amount of nanowire-based electric and photonic devices have been proposed and primitively demonstrated in the experiments. In the meantime, scientists are devoted to
studying the abundant physics behind in order to improve the performance and practicability of the devices. Here we introduce several examples of photonic devices hingeing on photon emission from nanowires and some highly relevant fundamental results, which motivate the original research in this thesis. More weight will be put on InP nanowires, which are used in our experiments discussed below.

The first example to be discussed is the nanowire laser. Interestingly, observation of lasing from ZnO nanowire in 2001[15] is commonly regarded as the milestone initiating the worldwide research in nanowire photonics. The unique characteristic of nanowires when used as lasers is that nanowire themselves could play a dual role as optical gain media and nanocavities at the same time. On one hand, being semiconductor material, when properly pumped either optically or electrically[18], stimulated emission can be achieved in the nanowires by recombination of electrons and holes. On the other hand, due to their 1D geometry and high refractive index, sufficient light reflection can be expected at the end-facet and a Fabry-Pérot cavity can be established. With photonic emission and confinement combined, lasing has been observed for plenty of semiconductor nanowire materials. Depending on the bandgaps of the materials used to grow the nanowires, lasing wavelength can be tuned from near-infrared to ultraviolet[16][17]. Recently, owing to the taper-free and clean end-facet growth accomplished with SA-MOVPE, lasing has also been obtained with InP nanowires[19]. In addition, the farfield directionality of laser emission from the end facet of nanowires has been touched both theoretically[20] and experimentally[21]. It is shown that the specific farfield emission pattern depends on the lasing
modes and is generally less directional compared with conventional bulk semiconductor lasers.

Another class of nanowire-based devices which intrinsically relies on its light emission is nanowire light-emitting-diode (LED). By definition, a LED is a p-n junction diode which could emit light when electrons are injected externally by a bias voltage, a process called electroluminescence. Traditional LEDs made from bulk semiconductor materials have became widespread in the lighting market. In order to miniaturise their footprint, the possibility to fabricate LEDs with semiconductor nanowires has drawn much interest recently. In contrast with lasers in which an undoped nanowire is used, the nanowire used for LEDs is necessarily doped in order to form a p-n junction. The approaches to build p-n junctions with nanowires can be roughly divided into two distinct categories, namely crossed-nanowire junctions[22] and heterostructures[23][24]. While single nanowire LEDs are more suitable to be used as incoherent light source in integrated optical chips, nanowire arrays are promising to used as illumination and display applications. For the latter case, the far-field emission pattern has been investigated by J. Motohisa et al.[25]

Beyond conventional lasers and LEDs, nanowires have been explored to facilitate single-photon emission in the last decade as well. Unlike lasers and LEDs which have a statistical distribution in the number of photons emitted within a given time interval, single-photon sources, which stand out as crucial for studying the quantum properties of light and developing quantum information applications, are demanded to emit individual photons in a regulated stream[26]. A common scheme to fabricate a single-photon source is to incorporate a quantum dot into a semiconductor microcavity. In this configuration, quantum dots act as the atom-like quantum system which generate single photons while microcavities provides the necessary photonic environment to enhance and direct the emission and thus facilitate the subsequent experiments or applications. As mentioned before, nanowires are potentially competent to essentially play the same role of the microcavities. It has been reported by many groups that quantum dots can be easily embedded (self-assembled) in nanowires with precise position and size control during the growth of nanowires and decent single-photon emission can be obtained from these quantum dots through optical or electrical pumping[27][28][29]. Based on this quantum-dot-in-nanowire system, a couple of proof-of-concept single photon devices have been made[30][31]. An example is schematically shown in figure 1.3. Alongside the effort put into developing the functional devices, enormous work has also been
done to study the specific physics concerning the photonic properties of quantum-dot-in-nanowire systems, including their spontaneous emission dynamics[32] and out-coupling radiation pattern[33]. With a deeper understanding of the mechanisms behind, one can improve the efficiency of the system accordingly with finer designs and fabrication methods. In practice, the efficiency of a single photon source is usually defined as the probability of coupling a photon in relevant mode into the first optical collector (lens or fibre). Two parameters govern the total efficiency jointly: the $\beta$-factor which represents the ratio between the number of photons emitted to a relevant mode and to all possible modes and the extraction efficiency $\eta$ which is defined as the ratio between the number of photons collected by the optics and all the photons emitted. While the $\beta$ factor near unity can be virtually achieved, the extraction efficiency remains a challenging concern since the radiation out-coupled from nanowires is generally divergent[20]. It has been suggested that the emission can be more convergent when the nanowire is gradually tapered near the end-facet such that the out-coupling is adiabatic[34].

Overall, nanowires have been proved to be highly potential for applications in lasers, light-emitting diodes and single-photon sources. For these three devices, in particular for the single-photon source, obviously it would be extremely meaningful if the emission is highly directional, which mo-
tivated the research of this thesis. For the specific topic of directivity of emission from nanowires, a lot of work has been done previously. First of all, the directivity of single nanowire emission has been attributed to waveguided/leaky modes\cite{35}. Depending on the diameter, different waveguide modes are supported by the nanowires and selectively coupled, leading to different far-field emission pattern. For example, in the case of a vertical InP nanowire with 100nm diameter, the emission is favourably coupled to the TM$_{01}$ mode that is leaky for this diameter and emission wavelength, which results in a ring-like far-field pattern, as shown in figure 1.4\cite{42}. Secondly, the emission from a highly ordered nanowire array could bear particular directivity as well. In this circumstance the emission can be coupled to Bloch modes supported by the periodic structure and the nanowire array behaves as a quasi-two-dimensional photonic crystal\cite{36}. Moreover, by varying the dimension of the nanowires and the geometry of the array, the interplay between the single nanowire emission and the nanowire array emission can be manipulated and a wide-range control over the emission directivity can be achieved\cite{37}. However, what has not yet been reported is the emission feature from finite number (more than one) of nanowires close to each other, which is a different structure from the above-mentioned single nanowires or nanowire arrays. In this thesis, we will tackle this issue and discuss the directivity of emission from nanowire dimer, which is the simplest case of such multi-nanowire configurations.

1.4 Methods of research

In the research of this thesis, the emission directivity from nanowire structures are studied by numerical simulation and experiment. In the simulations, the finite-difference time-domain (FDTD) method is employed through the commercial software Lumerical FDTD solutions while the experimental measurements are performed by means of Fourier microscopy. In this section a general introduction to these methods is presented while details about how they are implemented will be given in the relevant chapters later.

As its name suggests, FDTD method is a grid based time-domain numerical technique widely used to solve Maxwell’s equations. During FDTD calculations, Maxwell’s equations are discretized and then solved in a leapfrog manner which means that the electric field $E$ and magnetic field $H$ in a certain spatial volume are calculated alternately in time sequence. While only electromagnetic field intensity in the near-field is calculated directly,
Figure 1.4: Emission pattern of a single InP nanowire with 100nm diameter[42]. The polar coordinate $\theta$ is shown to the left while the azimuthal coordinate $\phi$ is labelled inside the main panel. The vertical cut and horizontal cut through the center are shown in the right and top panel respectively.

Fourier microscopy is an emerging method which is powerful and versatile to investigate the directivity of the luminescence or light scattering of nanostructures. The core mechanism of Fourier microscopy is to image the back focal plane of an objective lens (obtained image is called Fourier image hereafter). As illustratively shown in figure 1.5, a lens will transform an incident plane wave with a certain direction, described by $(\theta_x, \theta_y)$ or $(k_x, k_y)$, to a paraboloidal wave and subsequently focus it to a diffraction limited spot with defined spatial coordinate $(x', y')$ in the back focal plane. For any wave packet, it can be expressed as a superposition of a series of plane waves with unique $k$ vectors. When incident into the lens, different plane-wave components in the wave packet will be focused to different points in the back focal plane. In this way, the field intensity at every point of back focal plane corresponds to the weight of a unique component in k-space, which could be translated to the intensity of emission to a certain angle.
In principle, the Fourier image provides enough information to reconstruct the emission pattern of the sample.

Figure 1.5: Schematic illustration of performing Fourier Transform with a lens. Plane waves with different k-vectors are focused to different points in back focal plane. The inclined dash line through the centre of the lens indicates the particular path along which the light is not deflected by the lens.

1.5 Outline of the thesis

During the project, the emission directivity of two distinguishable InP nanowire structures, being vertical nanowire dimers and horizontal single nanowire respectively, has been investigated. Both structures will be discussed in the thesis, which is organized as follows:

In chapter 2 an introductory description about the waveguide modes supported by nanowires and their interaction with the light emission will be given. Afterwards, chapter 3 and chapter 4 will be dedicated to the discussion on the emission pattern of the vertical nanowire dimer and the horizontal single nanowire respectively. Finally, conclusions will be summarized in chapter 5.
Chapter 2
Theoretical Background

In this chapter we present some basic theories which are of the essence to discuss the emission property of our nanowire structures.

2.1 Waveguide modes in infinite cylinder

An infinitely long dielectric cylinder, sketched in figure 2.1 with relevant coordinates, is widely used as an ideal model for optical fibres. Although the length of a nanowire is far shorter and the cross section is generally non-circular, the waveguide modes supported by the infinite cylinder still form a good analytical approximation to calculate the modes existing in a real nanowire, considering its large aspect-ratio and azimuthal symmetry.

Figure 2.1: An infinite dielectric cylinder for calculating waveguide modes. The radius is $R$ and the refractive indices inside and outside the cylinder are $n_1$ and $n_2$ respectively.
Being a typical example of boundary value problem, the approach to obtain the waveguide modes in an infinite cylinder is fairly standard: solve the Maxwell equations with the boundary conditions imposed by the interface between the cylinder and the surrounding medium. Since we are only interested in propagating solutions in source free medium, the Maxwell equations can be reformed into a single wave equation (Helmholtz equation) with wave number \( k_0 = \frac{2\pi}{\lambda} \):

\[
\nabla^2 U + n^2(r)k_0^2 U = 0
\]

where \( U \) can be any component of electric and magnetic fields and \( n(r) \) is the refractive index, being \( n_1 \) in the cylinder and \( n_2 \) in the surroundings respectively.

In cylindrical coordinates, because of azimuthal symmetry, the component field can be written as \( U(r, \phi, z) = u(r)e^{-j\phi}e^{-jk_z z} \) with \( k_z \) being the \( z \)-component of the wavevector. Then wave equations leads to an differential equation for the radial profile of the field \( u(r) \):

\[
\frac{d^2 u}{dr^2} + \frac{1}{r} \frac{du}{dr} + (n^2(r)k_0^2 - k_z^2 - \frac{l^2}{r^2}) = 0
\]

(2.2)

After inserting the corresponding refractive index, the general analytical solution with several to-be-determined constants can be obtained for the space inside and outside the cylinder separately. The specific derivations are extremely tedious so we omit them here and readers are referred to the standard textbook of optical waveguide theory[38]. Subsequently, the boundary condition requires that the parallel field components are continuous at the interface, which can be expressed in cylindrical coordinates as:

\[
\begin{align*}
e_{1z}(R) &= e_{2z}(R) \\
h_{1z}(R) &= h_{2z}(R) \\
e_{1\phi}(R) &= e_{2\phi}(R) \\
h_{1\phi}(R) &= h_{2\phi}(R)
\end{align*}
\]

(2.3)

This set of conditions will help to determine the unknown constants existing in the general solution and yield definite expressions of the field profile. Meanwhile, they ultimately lead to the dispersion relation equation, solutions of which characterize the dispersion relations of different modes:

\[
\left[ \frac{\mu_1}{k_1 R} J'_1(k_1 R) - \frac{\mu_2}{k_2 R} H'_1(k_2 R) \right] \times \left[ \frac{\epsilon_1}{k_1 R} J'_1(k_1 R) - \frac{\epsilon_2}{k_2 R} H'_1(k_2 R) \right] = \left( \frac{k_z R}{\omega R/c} \right)^2 \left[ \frac{1}{(k_2 R)^2} - \frac{1}{(k_1 R)^2} \right]^2
\]

(2.4)
where \( J_l \) and \( H_l \) are the \( l \)th-order standard Bessel and first-kind Hankel functions. \( k_z = k'_z + i k''_z \) is the complex wavevector along the axis of the cylinder (\( z \) direction). \( k_i = [\epsilon_i \mu_i (\omega/c)^2 - k_{z_i}^2]^{1/2} (i = 1, 2) \) is the transverse component of the wavevectors inside(\( k_1 \)) and outside(\( k_2 \)) the cylinder, with \( \epsilon_i \) and \( \mu_i \) being the corresponding electric permittivity and magnetic permeability and \( n_i = (\epsilon_i \mu_i)^{1/2} \) being the refractive index. The modes for which \( k_2 \) is not purely imaginary are called leaky modes since they can radiate into the surrounding medium from the interface while the modes are guided in the cylinder otherwise. The propagation length is defined as \( L_d = 1/2k''_z \) to characterise the distance that the energy can flow in the cylinder. For leaky modes, the loss of energy confined in the cylinder is majorly caused by the outcoupling of light to the radiation zone through the side wall. Therefore the propagation length is also called leakage length in this case to indicate that it is the distance beyond which the energy is not mainly confined any more. For the guided mode, though the light cannot radiate out, the energy can be attenuated due to absorption. When inspecting the field profile, three different classes of modes can be defined, i.e. transverse magnetic modes (TM_{0m}) for which \( H_z = 0 \), transverse electric modes (TE_{0m}) for which \( E_z = 0 \) and hybrid electromagnetic modes (HE_{lm}). All the modes are labelled with two subscripts, \( l \) being the order present in equation 2.2 while \( m \) being the order of solution under the same \( l \). However, TM and TE mode only exist under the condition of \( l = 0 \).

For the specific case of an InP nanowire in air, the relevant constants are as follows: \( \mu_1 = \mu_2 = 1 \) since the media are non-magnetic; the InP refractive index is assumed to be \( n_1 = 3.42 \). With all the information at hand, the dispersion relations for all the modes can be obtained by solving equation 2.2. In figure 2.2 the dispersion relation, expressed as \( k_0d \) vs. \( k_zd \), are plotted for a couple of modes in the lowest order. When the wavelength \( \lambda (k_0 = \frac{2\pi}{\lambda}) \) is targeted at 870nm which is the emission peak of InP nanowire, a very important piece of information that can be extracted from the dispersion plot is the number and type of modes supported in the cylinder with a specific diameter. For example, when the diameter of the cylinder is smaller than 200nm, only the HE_{11} mode is guided.

### 2.2 Coupling of dipole emission with waveguide modes

As introduced in section 1.2, it is justifiable to simplify the emission of nanowire to a semi-classical scenario shown in figure 2.3, in which the emis-
Figure 2.2: Dispersion relation of leaky and guided modes supported in infinite long InP cylinder. The modes that lay in the light grey area are guided inside the cylinder and purely evanescent outside while those in the white area are leaky, which feature a non-negligible real part of $k_2$. As the dispersion relation is expressed as $k_0d$ vs. $k_zd$, the diameter is shown at the bottom horizontal axis for $n = 3.42$ and $\lambda = 870\text{nm}$.

Emission is considered to be from the electric dipole transition of electron-hole pairs. Therefore the emission is modelled as a dipole source and the presence of nanowire sets an inhomogeneous environment for the dipole, providing different waveguide modes (legitimate solutions of Maxwell’s equations). Upon emission, the photons could be coupled to certain modes, which ultimately influences the emission pattern. It has been proved that the coupling probability is proportional to the dot product of the dipole moment and the mode field intensity at the dipole position: $\vec{p} \cdot \vec{E}(\vec{r}_0)[39]$. The dot product decides that both the profile of the electric field and its direction are relevant for the emission coupling. Specifically, a certain mode will be favourably coupled and hence emission to that mode will be enhanced if its electric field is strongly confined in the area near the dipole source and the field component parallel to the dipole orientation is dominating. Similarly, the coupling could be inhibited when the product is minimal, either because the electric field is mainly perpendicular to the dipole or because the field intensity itself is minimal at the dipole position.
Figure 2.3: Illustration of light emission from nanowire. The radiation source is modelled as a dipole and the geometry of the nanowire is modelled as an cylinder. The emission from the dipole can be coupled to the waveguide modes supported by the cylinder.

As has been shown in figure 2.2, distinguishable modes are supported by a nanowire with a particular diameter. In general, depending on the orientation of the electric dipole model, the possibility of coupling emission to these different modes are not equal since they differ significantly in the field profile. As an example, the field profile for the TM\textsubscript{01} modes supported by a 100nm InP cylinder is plotted in figure 2.4. At the centre of the cylinder, the electric field component along the z-direction (parallel to the axis of the cylinder) is maximised while the component perpendicular to the axis is zero. Consequently, assuming only dipoles placed on axis are considered, the coupling between a dipole oriented along z-direction with TM\textsubscript{01} mode is favoured while it is virtually impossible to couple the emission from a dipole lying in the cross-section plane to TM\textsubscript{01}.

Figure 2.4: Field profile (expressed in Cartesian coordinates) of the TM\textsubscript{01} mode in an InP cylinder with 100nm diameter.
However, it should be noted that the discussion on the coupling probability above does not take the selection rules of the dipole transition into account, which are decided by the property of the material rather than the geometry alone. It has been reported that the crystal structure has a significant impact on the orientation of dipole model. Specifically, the zincblende (cubic) crystal structure allows for parallel and perpendicular dipoles with equal strengths, while the wurtzite (hexagonal) structure favours a perpendicular dipole[40].
Chapter 3

Emission Directivity of Vertical Nanowire Dimers

In this chapter we will discuss the far-field emission pattern of nanowire dimers, with a combination of FDTD simulations, Fourier microscopy measurements and a simple analytical model. The name nanowire dimer is coined to denote the structure of two nanowires situated close to each other with a typical distance of about one wavelength. As an analogous structure, the metallic dimer has been reported to be capable of directing light efficiently[43][44]. However, to our best knowledge, the nanowire dimer has remained unexplored before for emission directivity.

3.1 Simulations

Prior to any measurements, numerical simulations are powerful and necessary to provide some physical insight about the question being studied. In order to get briefed on the general radiation pattern of nanowire dimer structures and on how different parameters could affect the pattern, we performed a series of FDTD simulations with the commercially available software FDTD solutions of Lumerical Inc. Figure 3.1 depicts the general structure used in the simulations. First of all, the simulation region is set as a cuboid with its centre being the origin point. Only in this region the space is discretized according to the mesh accuracy setting and the field intensity is calculated at every mesh point. Finer mesh yields better accuracy but requires longer time. Based on some convergence tests, mesh accuracy 3 was chosen for the entire simulation region in pursuit of good trade-off. The Perfectly Match Layer (PML) boundary condition was employed for all the six sidewalls of the cuboid to avoid any reflection. Then the InP nanowire dimer was simplified as two cylinders and placed...
inside the simulation box with the centre of one at origin while another at \((x, y, z) = (d, 0, 0)\). The permittivity data of InP were taken from Palik[41], which is incorporated in the software. Finer cubic meshes with length of 9nm were used around the nanowires. The emission source was modelled as an electric dipole with a finite amplitude and phase which radiates at the wavelength of 870nm and positioned in the nanowire on the left-hand side. Finally, a 2D frequency-domain field monitor was introduced above the nanowire dimers to record the near-field intensity, from which the far-field emission pattern can be calculated after the simulation. In principle, it is required that the field intensity drops to zero when approaching the edge of the 2D monitor to ensure the accuracy of the near-field to far-field projection. Unfortunately this is not rigorously true in most situations. For the most precise results, one should perform the projection with a 3D monitor enclosing the simulation region. But this procedure involves a huge amount of data and is excessively time-consuming for a batch of simulations. To tackle this dilemma, we performed some trial simulations to compare the results from 3D monitor and from a series of 2D monitors with different sizes and positions. In this way, we believe that a 2D monitor placed at a proper position is able to give the far-field emission pattern with a sufficient accuracy therefore its use in the batch work is maintained.

Within the frame established above, a couple of parameters remain possible to be set freely, including the length and diameter of nanowires, the separation \(d\) as well as the polarization and position of the emitting dipole. Obviously it is not sensible to study all the variables simultaneously. For simplicity, the length of nanowires is chosen constantly as 3\(\mu\)m, and the diameters of the two individual nanowires are kept identical. In addition, the emission off the axis of the nanowire is not considered which means the dipole source is always placed on axis. While our primary concern is to
study the dependence of emission pattern on the separation $d$ of the two nanowire, all the other diameters were investigated in the simulations to a certain extent. Nevertheless, considering their complexity and relevance for guiding the measurement, they are studied with different emphasis. Discrimination is based on previous experiences from references and our first-hand results from simulations and will be explained below.

3.1.1 Nanowire Dimer with 100nm diameter

As discussed in chapter 2, diameter is determining parameter for the modes supported in the nanowires and thus their emission pattern. Since there are only a few modes supported by a nanowire as thin as 100nm, we started our simulations with this diameter. Grzela et al. have demonstrated that for a single nanowire of 100nm diameter, the emission coupled to TM$_{01}$ is dominating over HE$_{11}$ so that the emission pattern$^{[42]}$ is ruled by the leaky feature of TM$_{01}$. Since for the TM$_{01}$ mode the electric field inside the nanowire is predominately oriented parallel to the axis of nanowire, the radiation of a dipole source with its moment along the same direction is more intense compared with the dipole perpendicular to the axis. This fact is verified by our simulations, as shown in figure 3.2, where the emission pattern from a vertical dipole (along z-direction) and a horizontal dipole (along y-direction), both at the centre of a InP nanowire ($d = 100\text{nm}$), are compared. Clearly, the emission from the horizontal dipole under this geometry is much weaker. As a result, in the following simulations for the thin nanowire, the dipole source is fixed along the z-direction.

Another indefinite parameter is the position of the dipole source (z-coordinate in figure 3.1). The dependence of coupling emission to a certain mode on the emitter position has been thoroughly investigated in ref. 34. It is suggested therein that the coupling possibility to leaky TM$_{01}$ mode only weakly depends on the source position, providing the length of the nanowire is longer than the leakage length which can be approximated as around half micrometer in this particular diameter. We tested this inference with a single nanowire in a batch of simulation by placing the dipole source at five different positions, i.e $z = -1\mu\text{m}$, $z = -0.5\mu\text{m}$, $z = 0$, $z = 0.5\mu\text{m}$, $z = 1\mu\text{m}$, as schematically shown in figure 3.3 a). In figure 3.3 b) the farfield emission intensity in the x-z plane is displayed as a polar plot for every source position. On one hand, the emission patterns are qualitatively unchanged when the dipole source is placed at different point, sharing a typical symmetric two-lobe form in the cut plot (corresponding to a ring shape in the 2D colour map plot similar with the one shown in
Figure 3.2: Simulated emission pattern from a dipole source in the centre of a nanowire with 100nm diameter. a) dipole orientation parallel to the axis of the nanowire (z-direction) b) dipole orientation perpendicular to the axis of the nanowire (y-direction). The far-field intensity of both panels is normalized to the maximum value of the intensity in panel a). Note that the colorbar in panel b) is scaled as 1/10 of the one in panel a) in order to make the very weak emission in panel b) perceivable.

As will be discussed in section 3.2, our photoluminescence experiment is performed with pure InP nanowires and the emission region covers a relative large part of the nanowire. In order to avoid subjectively favouring a certain position in the simulations, we performed the simulation for every dipole position discussed above. In the case of 100nm diameter nanowire, given
the fact that the emission pattern from the dipole at different positions does
not deviate significantly from each other, we averaged the results from the
position dependent simulations and took the averaged pattern as the final
one for further discussion. However, from the viewpoint of comparing the
simulation with the experiment, even the averaging procedure involved
bias since the weights to be used for the different position is virtually
unknown and the same weight that we employed is rather arbitrary. Further
modification on experimental methods could circumvent this problem. For
example, one can limit the pumping area through cathodoluminescence
or otherwise limit the size of the emitting structure by incorporating a
quantum dot which emits at a different wavelength in the nanowire.

In figure 3.4 the averaged emission patterns from a single nanowire and
nanowire dimers with three different separations are shown together. Ob-
viously, with the introduction of an extra nanowire, the symmetry featured
by the emission pattern of a single nanowire is lost. Along the ring where
the emission from a single nanowire is strongest, the emission power is re-
distributed and considerably enhanced into certain angles. Remarkably,
the decrease of the separation distance leads to a smaller number of di-
rectional lobes where the emission is enhanced compared with the single
wire case. In particular, the emission from the dimer with 0.5µm separa-
tion is predominantly concentrated on the half space opposite to the extra
nanowire.
Figure 3.4: Emission pattern from single nanowire \((d = \infty)\) and nanowire dimers with different separation \(d\), which is shown on the top-right corner of each panel. All the data are normalized to a single maximum value. In the cases of nanowire dimers, the dipole source is positioned in the nanowire on the left-hand side, as depicted in figure 3.1.

3.1.2 Nanowire Dimer with 180nm diameter

In this section the nanowire with 180nm diameter is considered. A group of simulated emission patterns is displayed in figure 3.5 where the emission patterns of a single wire (top row) and a nanowire dimer 1\(\mu m\) apart (bottom row) are compared. In this geometry, the field profile of the HE_{11} mode becomes more confined and easier to be coupled to by a dipole in the nanowire while the TM_{01} mode is still available as well. For the electric field of the HE_{11} mode, the component perpendicular to the nanowire axis is dominant over the parallel component. Therefore, a horizontal dipole would be favourable to excite it. As a result, in the simulations for the nanowire of 180nm diameter, no assumption could be imposed on the orientation of the dipole source and thus all the three dipoles orthogonal to each other should be taken into account, unlike the case of the thin wire where the horizontal dipoles are neglected. However, practically the three orthogonal dipole sources should be incoherent whereas they would emit coherently.
Figure 3.5: Simulated emission pattern of single wire (top) and nanowire dimer with separation $d = 1\mu m$ (bottom). Different columns correspond to different dipole positions on the axis of the nanowire, as indicated above each column.

in the simulation program if they are simulated in a single run. Therefore we need to perform the simulation individually for every dipole orientation and add up the emission from the three sources incoherently afterwards. The weights of the three terms for this summation procedure are arbitrarily taken as the same here. Moreover, at the diameter of 180nm, the coupling probability to the $HE_{11}$ and $TM_{01}$ modes is far more strongly dependent on the dipole source position[35]. Consequently, we simply retain the original patterns obtained from every batch of simulations performed for a specific position of dipole.

A couple of messages could be inferred from figure 3.5. First of all, the emission pattern indeed varies dramatically when the dipole source is moved from one point to another. The reason for this variation is lying in the selective coupling of dipole at different positions to different modes. A favourable coupling to $TM_{01}$ gives a ring-like emission pattern, as is the case for $z = -0.5\mu m$ and $z = -1\mu m$, while the emission is concentrated in the forward direction when $HE_{11}$ is coupled by the dipole at $z = 1\mu m$. Plausibly, an relatively equal coupling to both $TM_{01}$ and $HE_{11}$ in the particular case of $z = 0.5\mu m$ makes the emission pattern bear more complex features. Secondly, the influence on the emission from the extra nanowire is noticeable only for large angle emission which is typically from the emission
coupled to TM\textsubscript{01} mode. For the emission propagating towards the forward direction the difference is not very pronounced.

3.2 Measurements

The simulations discussed above provide a clear indication that introducing an extra nanowire close to an emitting one can change its emission directivity and get the emission power focused on a smaller solid angle. In this section Fourier microscopy measurements on the nanowire dimer are discussed.

3.2.1 Sample description

The nanowires used in the measurement is grown on top of an InP substrate with the selective-area metal-organic-vapor-phase-epitaxy methods. As described in section 1.3, hole arrays with a certain pattern are etched in a layer of mask film with e-beam lithography ahead of growth and afterwards the nanowires are grown at the positions of holes with the same diameter of the etched holes. The designed pattern for our sample is shown in figure 3.6 and an dark-field optical microscopic image is displayed in figure 3.7 for the specific array of dimers with 1\(\mu\text{m}\) separation. In order to eliminate the modulation on the emission pattern from the periodic structure, the pitch of the array is designed as 5\(\mu\text{m}\), which is too large for the Bloch modes supported by the array to exert influence. In other words, the emission collected in our experiment is guaranteed to be only decided by the single nanowire or dimer which is excited while the scattering from the surrounding nanowires in the array is negligible.

As has been shown in the simulations, the emission pattern from the leaky TM\textsubscript{01} mode is more liable to be modified by an extra nanowire. And in the case of thin nanowires (100nm diameter) TM\textsubscript{01} is the dominant mode being coupled to. With this knowledge, we only investigated the 100nm one in our measurement despite the fact that nanowires with five different diameters are arranged together in our sample.

3.2.2 Experiment procedures

The Fourier microscopy measurements are performed with a set-up shown schematically in figure 3.8, which is consist of three modules, i.e a sample stage, an optical Microscope and an acquisition box. During the experiment, the sample is placed on a 3D movable stage whose position can be
Figure 3.6: Schematic design of the sample pattern. In terms of dimer separation, four cases are included, with the specification shown on the top. In each cluster with the same dimer separation, there are five arrays for which the diameter of the etched holes is varied from 100nm to 300nm, as indicated in the arrays of single holes.

Figure 3.7: Dark-field optical microscopic image of a dimer array with 1µm separation. Because of the imperfect growth, considerable inhomogeneities in size of nanowires are present.
Figure 3.8: Schematic diagram of the Fourier Microscope

precisely controlled. Right above the sample stage there is a commercial
Leica microscope equipped with an objective, which is the key component
of Fourier microscopy (see section 1.4). Through the objective, a beam of
640nm laser light is focused on the sample to excite the photoluminescence,
which is collected by the same objective subsequently. In this way, the size
of the laser spot on the sample is believed to be confined to the diffrac-
tion limit. The objective used in this project is under the specification of
magnification factor 100× and numerical aperture 0.95, which guarantees
that the set-up is capable of collecting the light with polar emission angle
from 0° to 72°. A CCD camera (CCD Real in figure 3.8) is installed in
the microscope module to capture the real image of the sample and fa-
cilitate the focusing. In the acquisition module, the emission is divided
by a beam splitter to two paths. Through the first path the emission is
coupled to a spectrometer, by which the emission spectrum is recorded. A
typical photoluminescence spectrum when the pumping laser is focused on
a nanowire is shown in figure 3.9. Both emission from the nanowire and
from the substrate, which are fitted separately with Lorentz curves, can be
perceived. The emission peak from the InP nanowire, which has primarily the wurtzite crystal structure, is around 870nm. It is blue-shifted from the 910nm emission of InP zincblende substrate. Another path leads to a CCD camera (CCD Fourier in figure 3.8) to image the back focal plane of the objective. A lens (named with Fourier lens) is placed in the middle of the optical path between the back focal plane and the CCD sensor, at distance of its focal length from both, to construct a 2f system. In such a way, the Fourier image is projected on the CCD with its size unchanged. Since we are specially interested in emission pattern of the nanowire rather than the substrate, a 870nm band-pass filter (10nm bandwidth) is installed onto the Fourier CCD camera to guarantee that the predominant contribution for the Fourier image is from the nanowire. Additionally, because the transmission of the beam splitters considerably depends on the light polarization, only the Fourier image obtained with polarized light is absolutely physical. In order to get a correct unpolarized Fourier image for our nanowire emission, we need to take two orthogonally polarised images first, and then add them up with a correction ratio. The way to get the ratio is to carry out the same procedures for the substrate emission and search for a relative weight that could equalize the two orthogonally polarized images in intensity. The rationale for this correction methods lies on the fact that the emission from the substrate is supposed to be polarization independent[45].

3.2.3 Results

In this section we present a couple of Fourier images obtained for different nanowire dimer structures and attach the relevant simulation results alongside as well. But due to the specific working principle of the Fourier microscopy, the Fourier images obtained experimentally are not exactly the same as the emission pattern expressed in azimuthal coordinates. Instead, they are naturally expressed in k-space. Therefore the simulation results shown in this section are transformed from azimuthal coordinates to k-space (Cartesian coordinates) for convenient comparison. Moreover, as described above, the numerical aperture of the objective used in our measurement is 0.95, which means that only the emission under the condition of $k_\parallel/k \leq 0.95$ ($k_\parallel = \sqrt{k_x^2 + k_y^2}$) can be collected. Thus the simulation results are also shown in the same range.

We start our discussion with the single nanowire emission as the reference, which has been investigated in ref.40. In figure 3.10, the simulated and experimental Fourier image for the emission from a single 100nm
Figure 3.9: Emission spectrum recorded from the spectrometer. Lorentz fit is used to distinguish the peak from the InP nanowire and the peak from InP substrate. The grey dash line indicates the 870nm band pass filter used to only capture the emission from the nanowire.

nanowire is shown together. As expected, the pattern of emission is azimuthally symmetric and the intensity maximum exists at a quite large $k_\parallel$. However, while the results shown in ref. 40 bears a better resemblance with the simulation result, the result obtained for our own sample shows a more conceivable discrepancy. Firstly, the intensity contrast between the maximum and minimum in our case is not very high. Secondly, the intensity maximum appears at a larger $k_\parallel$ in the measurement result b) than the one in simulation result a). The possible reason for the former is that the contribution of emission from the substrate can be slightly higher than expected, which weakens the contrast as the substrate behaves as a Lambertian source and the emission intensity is highest in the forward direction. With respect to the second difference, there are two plausible causes. In the first place, the diameter of the grown nanowires might be thinner than 100nm. Another one could be that the coupling to the TM$_{01}$ mode is not extremely efficient due to the quality of the nanowire and emission from a free dipole is stronger than expected, in which case the the emission mainly radiates to the direction of $k_\parallel/k = 1$.

Next we proceed to discuss the Fourier image of the emission from nanowire dimers. In figure 3.11, the results of a dimer with 0.5µm separa-
Figure 3.10: Fourier image of the emission from a single nanowire with 100nm diameter. a) simulation result b) experiment result.

Figure 3.11: Fourier image of the emission from a 100nm nanowire dimer with separation of 0.5µm. a) simulation result b) experiment result. In both case the primary emission is arranged to be from the nanowire on the left-hand side of the dimer, as schematically shown on top of the graphs.
tion are shown in the same way as used for single wire in figure 3.10. Note that in the simulation the dipole source is placed in the nanowire on the left-hand side while in the experiment the pumping laser is also focused on the nanowire at the same relative position. In this way, the primary emission is just (predominantly in the experiment) from that nanowire. Clearly, the unsymmetrical emission pattern demonstrated in the simulation is observed to some extent in the experiment as well. And as expected, more emission goes to the half space that is opposite to the non-emitting nanowire. Unfortunately, when being pumped during the measurement, the nanowire on the right-hand side of the dimer did not emit such properly that no Fourier image with decent contrast was obtained for it. In figure 3.12, we display the Fourier image for a pair of nanowire dimer with 1µm separation. In this case the nanowires on both left-hand side and right-hand side can be efficiently excited and emit sufficiently. As shown in figure 3.12 b) and c) respectively, the emission pattern when the nanowire on the left is excited is opposite to the one when the nanowire on the right is excited, with the direction consistent with our anticipation.

Although the similarity between the simulation results and the experimental results is fairly clear, there do exist some discrepancies. Compared with the simulation settings, the most noteworthy difference in the experiments is the existence of the substrate. Apart from the fact that the emission from the substrate itself could blur the contrast as mentioned before, it could also modify the emission pattern of a dipole positioned closely on top of it[39]. Additionally, the nanowires being investigated in the experiments might be geometrically imperfect, which could compromise the obtained Fourier images. Since it is difficult to estimate definitely the effect imposed by these factors, the comparison must be treated with caution.

3.3 Analysis with a simple model

By a combination of simulation and measurement, we have shown that a nanowire dimer show a different emission pattern with a single nanowire. As apparent from the results shown earlier, depending on different separation of the nanowires, the emission get enhanced at different angles in farfield. In general, more emission power flows to the direction opposite to the nanowire that is not emitting (not pumped). We believe this particular emission directivity is due to the interference of the emitted light from the emitting nanowire and the scattered light from the non-emitting nanowire.
Figure 3.12: Fourier images of emission from InP nanowire dimer with 1\(\mu\)m separation. a) simulation result with the dipole source in the nanowire on left-hand side. b) experiment result with excitation at the nanowire on the left-hand side. c) experiment result with excitation at the nanowire on the right-hand side.

To gain a deeper insight of the physics behind the directivity, we conceived a simple dipole-dipole model, as sketched in figure 3.13, in which the nanowires are simplified as dipoles and their emission is observed at a farfield position \(A\). Consistent with the treatment in the simulations and the experiments, dipole \(p_1\) models the emission of the nanowire pumped by the laser while dipole \(p_2\) models the scattering of the non-emitting nanowire. Obviously, the two dipole sources are coherent since the oscillation \(p_2\) is driven by \(p_1\). The rationale for this simplification lies in two facts. First of all, the dominant mode in the thin nanowire (100nm) is the leaky TM\(_{01}\) mode, which means that the farfield profile of the emission is only modestly changed from the original emission of a vertical dipole. Secondly, the scattering field of the other nanowire, though rather complex, is always possible to be expressed in multiple expansion. For the first order approximation, only the dipole term is maintained. Therefore, in the simple model, both the primary emission and the secondary scattering are simplified as dipole sources.
Figure 3.13: A simple analytical model for the nanowire dimer emission. The nanowires are simplified as dipoles. Dipole $p_1$ located at origin models the emitting nanowire while dipole $p_2$ placed at $(d, 0, 0)$ models the scattering nanowire.

Within the model, the relevant parameters are as follows: the amplitudes of the two dipoles are $|p_1|$ and $|p_2|$ respectively; the distance between the dipole positions is $d$, which is assumed to be the same as the separation of the two nanowires; the phase delay between $p_2$ and $p_1$ is denoted as $\Delta \psi_0$. At a farfield point $A(\theta, \phi, r)$, due to the fact that $r \approx r' \gg d$, the electric intensity vectors generated by the two dipole sources are approximately parallel to each other, both along the direction of $\hat{\theta}$. They can be expressed as [6]

\[
\begin{align*}
\vec{E}_1(\vec{r}, t) &= -\frac{\mu_0 |p_1| \omega^2}{4\pi} \cdot \frac{\sin \theta}{r} \cdot e^{i\omega t} \hat{\theta} \\
\vec{E}_2(\vec{r}, t) &= -\frac{\mu_0 |p_2| \omega^2}{4\pi} \cdot \frac{\sin \theta}{r} \cdot e^{i\omega t + \Delta \psi_0 + \Delta \psi_{\theta, \phi}}
\end{align*}
\]

Clearly, the difference only exists in the phase term. The first difference comes from the intrinsic phase delay $\Delta \psi_0$ which is not dependent on the observation position but intuitively decided by the separation $d$. The second term $\Delta \psi_{\theta, \phi}$ is introduced because of the different optical paths between the point $A$ and two dipole positions, which obviously depends on the coordinates of point $A$. It can be proved that

\[
\Delta \psi_{\theta, \phi} = -kd \sin \theta \cos \phi
\]
Simply taking the vectorial addition of the two part in equation 3.1 yields the total field intensity at A:

\[ \vec{E}(\vec{r}, t) = -\frac{\mu_0 \omega^2}{4\pi} \cdot \frac{\sin \theta}{r} e^{i\omega t} \left( |p_1| + |p_2| e^{i(\Delta \psi_0 - kd \sin \theta \cos \phi)} \right) \] (3.3)

And finally for inspecting the emission pattern, the time averaged Poynting vector can be expressed as

\[ \langle \vec{S}(\theta, \phi, r) \rangle = \frac{|p_1|^2 \mu_0 \omega^4}{16\pi^2 c} \cdot \frac{\sin^2 \theta}{r^2} \cdot (1 + 2\eta \cos(\Delta \psi_0 - kd \sin \theta \cos \phi) + \eta^2) \] (3.4)

where a new parameter \( \eta \) is defined as \( \eta = |p_2|/|p_1| \) which intuitively should be a number no larger than unity. With this treatment, only two parameters, namely \( \Delta \psi_0 \) and \( \eta \), are left for fitting if we are only interested in the emission pattern. While the emission pattern is fundamentally dependent on the phase delay \( \Delta \psi_0 \), it is tested to be less sensitive to the ratio \( \eta \).

Figure 3.14: Emission patterns of the analytical model shown in figure 3.13. The ratio \( \eta \) in all panels is chosen as one. a) Emission pattern of a single dipole b) Emission pattern of two dipoles separated by 0.5 \( \mu m \). the phase delay is chosen as \( \pi/2 \) c) Emission pattern of two dipoles separated by 1 \( \mu m \), the phase delay is chosen as \( 5\pi/4 \).

In figure 3.14 we display the emission patterns of three cases based on equation 3.4. Obviously, due to interference, emission patterns of the two coherent dipole sources are considerably different from the single dipole emission. Remarkably, to a certain extent they resemble the emission patterns of the nanowire dimers, which strongly suggests that the particular emission directivity of nanowire dimers indeed results from the interference of the emitted and scattered light.

### 3.4 Outlook

The discussion in this chapter thus far has demonstrated the possibility to change the emission directivity of a thin nanowire by constructing a
dimer with an extra nanowire. And we believe that the mechanism lies in the farfield interference between the light emitted by one nanowire and its coherent light scattered by another nanowire. With this understanding, we can envision that it is possible to tailor the emission pattern and improve its directionality by employing the tunable interference pattern with nanowire ensembles that are more complex than a simple dimer. Note that the interference is only a process of spatial redistribution of energy in which the energy is not dissipated. This particular character means that the energy loss will not be considerably enhanced when more nanowires are introduced, which is significant from a practical point of view.

Figure 3.15: a) A multi-nanowire structure. The emitting nanowire is positioned at the centre and five scattering nanowires are positioned evenly along a semicircle (r=0.5µm) surrounding the nanowire in the centre. All the nanowires are in the diameter of 100nm. b) Simulated emission pattern for the structure shown in a)

Though beyond the main focus of the thesis, we include a simulated emission pattern from a multi-nanowire (D = 100nm) structure in figure 3.15 to evidence our prediction about the possibility to get more directional emission from properly designed nanowire ensembles. The specific structure is sketched in figure 3.15 a), where one emitting nanowire is positioned in the centre and five scattering nanowires are positioned evenly along a semicircle. The distances from the surrounding nanowires to the centre one are 0.5 µm. The simulated emission pattern shown in b) suggests that the emission from such a structure is notably concentrated into one lobe towards the direction of the opening of the semicircle (opposite to the ensemble).
Chapter 4

Emission Directivity of Horizontal Single Nanowire

While in chapter 3 the directivity of nanowire dimers vertically standing on a substrate has been discussed, a different configuration—a nanowire horizontally lying on a substrate—is touched in this chapter.

Many nanowire-based experiments and devices have been achieved with a nanowire lying on a certain substrate\cite{15}\cite{24}. In such a configuration, it is more convenient to connect the nanowire with nano electrodes to perform electrically driven measurements\cite{18}, or incorporate the nanowire into hybrid structures to control its emission\cite{46}.

Following the same story introduced in chapter 2, in the horizontal nanowire, the emission is capable as well to be coupled to the waveguide modes supported by the nanowire. Depending whether it is a guided mode or leaky mode, the emission will be coupled out to radiation zone through the side facet or end facet of the nanowire. In this chapter we will show that the emission directivity under this configuration is significantly dependent on the source position. We believe that this result is relevant to improve the efficiency of the devices such as nanowire LEDs and nanowire-based single photon sources when a horizontal nanowire configuration is employed.

4.1 Measurement

The sample used in this measurement consists of horizontal InP nanowires lying on a layer of SiO$_2$ substrate. The InP nanowires were initially grown with the VLS method and subsequently transferred to the substrate. A
SEM image (view from above) for the particular nanowire that is investigated here is shown in figure 4.1 a), from which it can be measured that the length of the nanowire is $5.4 \pm 0.1 \mu m$ and the diameter varies from $190 \pm 5 nm$ (bottom side) to $245 \pm 5 nm$ (top side). Figure 4.1 b) displays the emission spectrum of the nanowire in a). As the substrate material in this case is SiO$_2$, only photoluminescence from the InP nanowire is observed when pumped with the 640nm laser. Though simply fitted with a Lorentz curve in figure 4.1 b), the emission spectrum shows small fluctuations around the main peak, which suggests the establishment of the cavity mode because of the reflection of the guided light at the end facets of the nanowire[47].

![SEM image](image1.png)  ![Emission spectrum](image2.png)

Figure 4.1: a) SEM image of the horizontal nanowire lying on SiO$_2$ substrate. Scale bar indicates 1$\mu$m. b) Emission spectrum of the nanowire shown in a)

Due to the horizontal orientation of the lying nanowire in reference with the substrate, it is practically possible to study the dependency of its emission pattern on the source position. The approach to achieve this within the frame of our Fourier microscope set-up is schematically illustrated in figure 4.2 a). With the help of the real camera mentioned in figure 3.8, we can focus the pumping laser into different parts of the nanowire. In this way, only the part being illuminated, the length of which is supposed to be comparable to the wavelength of the laser (640nm), can be excited. Then the emission source is effectively confined to the same region, considering the fact that the diffusion length of the carriers (electrons and holes) is
typically restricted within 100nm\cite{48}. The Fourier images obtained when the source position is in the middle, top or bottom part of the nanowire are shown in figure 4.2 b), c) and d) respectively. Note that since in this configuration the emitted light is highly linearly polarised along the orientation of the nanowire, a linear polarisation along the same direction was used in the measurement. Obviously, the emission directivity is indeed critically dependent on the source position. Whereas the emission goes to both directions when the middle part of the nanowire is excited, the light is predominantly coupled out to one direction when the excitation is near the end facet. And the favourable emission direction consistently corresponds to the direction from the middle to the excitation point, which suggests that the out-coupling of the light is occurred at the end facet nearer to the source point.

Figure 4.2: Source position dependent emission pattern of the horizontal nanowire. Sketch of exciting different part of the nanowire lying on a substrate with a pumping laser is shown in panel a). The Fourier images are given in b), c) and d), with the corresponding excitation positions indicated in a). Note that for this measurement a linear polariser along the orientation of the nanowire was used.
4.2 Analysis and Conclusions

As the diameter of the nanowire investigated here is about 220nm, in principle there exists three modes $\text{TM}_{01}$, $\text{TE}_{01}$ and $\text{HE}_{01}$, which can be concluded from figure 2.2. Considering the facts that the field profile of $\text{HE}_{11}$ in this diameter is more confined than the other two [35] and that the wurtzite structure of the nanowire favours a dipole oriented perpendicular to the axis, we think that the emission is primarily coupled to the $\text{HE}_{11}$ mode in this case.

Figure 4.3: Emission pattern obtained when different parts of a thin nanowire ($D = 100\text{nm}$) are excited. For emission pattern, the corresponding excitation position is indicated in the sketch above it. Again, a linear polariser along the orientation of the nanowire was used in this measurement as well.

With only a single measurement, admittedly it is difficult to draw any sound conclusions. So in order to build a deeper understanding on our results, we take advantage of some unpublished work done before within our group and our collaborative theory group. First of all, in figure 4.3 we show the excitation position dependent emission pattern of a lying thin nanowire ($D = 100\text{nm}$) obtained experimentally with Fourier microscopy. With the corresponding excitation positions being indicated in the sketch shown above, it is revealed by the emission patterns that in this case the light is mainly emitted to the opposite direction when the nanowire is excited near an end facet (left end excitation leads to a rightward emission and vice versa). This particular directivity is virtually opposite to the one shown in the thicker lying nanowire discussed before. For the diameter of
100nm, as discussed thoroughly in chapter 2 and chapter 3, the dominant mode being coupled to is the leaky TM$_{01}$ mode.

The contrary emission directivity from a thin and thick horizontal nanowire when they are excited at the end suggests that out-coupling character of different modes bears different source position dependency. Or stating it in the other way around, the specific modes being coupled to decides the direction where the emission radiates. This deduction is corroborated by FEM simulations performed by our collaborators from CSIC in Spain, which is shown in figure 4.4.

Figure 4.4: Simulated emission patterns from a lying nanowire with diameter $D = 100\text{nm}$ and length $L = 3\mu\text{m}$. The specific dipole source settings for every pattern is shown on its left, where $L_{ov} = 300$ indicates the length of emitting region (blue area in the nanowire).

The diameter of the nanowire investigated in this simulation is 100nm, which means the coupling between the emission from a dipole parallel to the nanowire and the TM$_{01}$ mode is dominant, as solidified again by the huge difference between the emission intensity in a), b) and the one in c), d). What is relevant here is that the simulated emission patterns also reveal the source position dependency of the emission pattern, as can be seen by comparing the pattern in panel a), c) and the one in panel b), d). More importantly, though both placed near the bottom end, the dipole
Figure 4.5: The out-coupling mechanisms for a) the guided HE$_{11}$ mode and b) the leaky TM$_{01}$ mode.

oriented perpendicular to the nanowire in panel d) leads to downward light emission while the dipole along the nanowire in panel b) results in upward light emission. Recall the fact that a perpendicular dipole is more likely to get coupled to the HE$_{11}$ mode while a parallel dipole tends to couple with the TM$_{01}$, even though the coupling efficiency of the former is much weaker than the latter in such a thin nanowire. So it is verified from the simulation that depending on whether it is coupled to the TM$_{01}$ mode or the HE$_{11}$ mode, opposite directivity of out-coupling emission could be obtained when a dipole source is located near the end of a lying nanowire.

With respect to the physical reason for the findings, our supposition is that the out-coupling mechanisms for different modes are not the same, which are schematically illustrated in figure 4.5. Specifically, for the the HE$_{11}$ mode which is guided regardless of the diameter of the nanowire, it is only possible to be coupled out from the end facet of the nanowire to the radiation zone. The out-coupling efficiency (transmission at the interface) is supposed to be fairly efficient thus a significant portion of the emission incident on the nearer end facet gets coupled out while the emission energy towards the further end facet is considerably dissipated inside the nanowire and the outcoupled radiation to that side becomes very weak. In contrast, in a thin nanowire, the TM$_{01}$ mode is leaky, which means that the light could radiate out through the side wall of the nanowire. However, if the travelling length for the light in TM$_{01}$ is less than its leakage length as defined before, the power flow will be mainly confined in the nanowire like a guided mode. When the nanowire is excited near a end facet such that the distance between the emission source and the nearer end facet is shorter than the leakage length, essentially no emission originally directed towards the nearer end facet could be coupled out through the side wall before reaching the facet. And apparently it is not very efficient for the light in
the TM$_{01}$ mode to be coupled out at the end facet interface. Therefore, the light is predominantly reflected by the nearer end facet and coupled out through the side wall afterwards, which explains that the out-coupling emission mainly goes towards the direction opposite to the nearer end facet.
Chapter 5

Conclusions

In conclusion, we have discussed in this thesis the particular emission patterns of two nanowire based structures, by means of simulations, Fourier microscopy measurements and theoretical analysis.

Firstly, the emission pattern from the dimer structure made up of two identical nanowires with 100nm diameter is demonstrated to be noticeably different with the emission pattern of a single nanowire with the same dimension. In this case, more power is radiated into the direction opposite to the non-emitting nanowire and the emission is more directional compared with the single nanowire emission. We attribute the mechanism for this emission pattern to the farfield interference between the originally emitted light by one nanowire and the scattered light by another nanowire in the vicinity. Potentially, well-designed structures with more complex patterns could be used to improve the directionality of the emission further, with the mechanism remaining essentially the same.

Secondly, the emission pattern from a horizontal nanowire lying on a substrate is proved to be source position dependent. While the emission pattern is basically symmetric when emission source is located in the middle of the nanowire, the light is disposed to radiate towards a certain direction when the source is moved close to the end facet. Interestingly, the particular correlation between the emission directivity and the source position depends on the modes that are coupled to, which hypothetically results from the different out-coupling paths of the leaky and guided modes.
Bibliography


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