Characterization of Single Emitters and Nano-Antennas.
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Summary

A main promise of the field of physics called “nanophotonics” that deals with nanotechnology to control light, is to provide control over the emission of light sources. Recent developments in the field of solid state lighting that resulted in bright and efficient LEDs, as an example, rely directly upon our understanding of how to control the emission of light. On the other hand, scientific advances in the fields of super-resolution microscopy, optical and quantum information processing, cryptography and secure communication, significantly rely on advances in controlling the emission of single photons, the detection of single photons, and increasing the light-matter interaction strength between emitters and single photons. In all these applications, the main question is how to control the emission of light in terms of the rate at which the source emits, the direction into which it radiates, and the polarization in which the source emits a photon. In this thesis we address how to choose an efficient single photon emitter, how to characterize a nanophotonic structure which controls the rate and direction of the emitted light, and how to fully characterize the polarization of the emitted light.

In chapter 2, we study the statistical properties of single nanodiamond NV centers. Specifically, we quantify the brightness and the decay rate of many individual NV centers in two sets of diamond nanocrystals with mean diameters of 25nm and 100nm. Our quantifications show that for both sets of nanodiamond diameters, the brightness and the decay rate of the NV centers are widely distributed, a finding that is in agreement with previous reports. The common opinion in the field so far has been based on the speculation that this distribution is entirely due to size dependent variations of the ‘local density of optical states’ in different diamond nanocrystal matrices, while assuming a near unity quantum efficiency for the NV centers. In contrast, based on the absence of strong correlation between the measured decay rate and brightness of the NV centers, we suggest that the wide distributions in decay rate and brightness are likely due to a distribution in the quantum efficiency of the NV centers. To support this, we calculate the local density of optical state variations according to the size distribution of our nanodiamonds and we find that it is not sufficient to describe our measured decay rate distributions and we argue that there should hence be a wide distribution in both radiative and nonradiative decay rates and, therefore, also in the quantum efficiency of the nanodiamond NV centers.

Our findings in chapter 2, showing that the quantum efficiency of NV centers in nanodiamond does not correlate with their readily measurable quantities like spectrum and brightness is crucial to nanophotonics application where NV centers are used as efficient fluorescent emitters or probes of the photonic environment. This implies that one can not choose the best NV center only based on its brightness, spectrum or decay rate. Instead one needs to utilize experimental protocols to directly quantify the quantum efficiency of each individual nanodiamond NV center before applying it to the target nanophotonic assembly as an emitter or probe. We introduce such protocols in the next chapter.

In chapter 3, for the first time, we quantify the quantum efficiency of individual nanodiamond NV centers reversibly and nondestructively. To this end, we use two techniques to apply known variations in

the local density of optical states to the photonic environment of the NV centers. In the first method, we vary the LDOS using liquids with different refractive indices which we apply to the 25nm nanodiamonds. We find that the small nanodiamonds are insensitive to the moderate variations of the LDOS. In connection with the measured decay rate distribution of the NV centers in chapter 2, we conclude that the 25nm nanodiamond NV centers have a distributed low quantum efficiency between 0% and 20%. In the other technique, we use a metal coated micro-mirror bead attached to the end of a near-field scanning tip to induce known LDOS variations. This is realized by nanomechanically changing the distance between the mirror and individual NV centers. Using this technique with 100nm nanodiamonds, for the first time, we report the results of quantitative measurements of the quantum efficiency of single NV centers. We show that the quantum efficiency of nanodiamond NV centers can be far from unity and is indeed widely distributed between 10% and 90%. This is in agreement with our assumption in chapter 2 and in contrast to the common belief that considers a near unity quantum efficiency for all nanodiamond NV centers. This highlights the importance of utilizing a nondestructive and reversible quantum efficiency calibration method, as shown in this chapter, to screen nanodiamond NV centers as efficient emitters or probes.

While chapters 2 and 3 involve how to choose the best emitter, in chapter 4, we discuss how to control and characterize the emission properties of such an emitter when it is coupled to a nanophotonic structure. Specifically we study a new type of nanophotonic antenna known as plasmonic patch antenna which consists of a circular patch of metal that is separated from a metallic ground plate via a dielectric material of several 10nm thickness. This antenna has already been shown to enhance the spontaneous emission of the emitters that are coupled to it while providing a very directional emission. In this chapter we use a technique called angle-resolved cathodoluminescence imaging spectroscopy (ARCIS) to characterize the spatial modes and emission properties of the plasmonic patch antennas. This technique is based on scanning an electron beam over the antenna structure to excite the plasmonic modes of the antenna. By collecting the induced and scattered light, we obtain high-resolution maps of the spatial and spectral mode structure, as well as the angular emission patterns of the plasmonic patch antenna. To understand the observed modes, we use a semi-analytical model that is based on LDOS variations in the patch antenna. We find qualitative similarities between the experimental and the calculated maps. However a one-to-one quantitative agreement is not present which we assign to the complex nature of mode excitation by the electron beam. Furthermore, we present the results of the measured angular distribution of the antenna radiation and find that for resonant excitation of the antenna, the emission is highly directional with a doughnut-shaped beam profile. Although this beam profile has been predicted in previous reports, it has not been verified experimentally. Moreover, we observed directional beam steering in the patch antenna which we explained using a simple distributed dipole model, where, the scattering of the light (surface-plasmon-polaritons) at the antenna edges are modeled by a set of radiating dipoles. We find good agreement between the model and the experimental radiation patterns of the antenna at different excitation positions.

Finally, in chapter 5, we present a new microscopy technique to fully map the polarization state of the light scattered or emitted by a single nano-object for all the different directions in its angular radiation pattern. We realize this by using a Fourier microscope that images the back focal plane (k-space) of a

high-NA objective, in combination with a Muller polarimeter, which consists of a polarizer and a quarter-wave plate. We apply our technique to the fluorescence of dye molecules residing in the central nanoaperture of so-called plasmonic bull's eye antennas, and also in spiral antennas which consist of concentric or spiral metallic grooves around the nanoaperture. Using our technique, we characterize the emission from the bull's eye and the spiral plasmonic antennas in terms of the total degree of polarization as well as the degree of circular and linear polarization. Moreover, we extract the orientation and eccentricity of the polarization ellipse for the polarized component of the emitted fluorescence light from our antennas. We show that the emission from both antennas display a significant degree of polarization, while the emission from a simple nanoaperture lacks any strong degree of polarization. We assign the observed degree of polarization to the fact that a part of the fluorescence emitted by the molecules is captured by surface plasmon polaritons, that subsequently radiate out in a directional manner. Since surface plasmon polaritons are p-polarized surface waves this indirect, plasmon-mediated, emission becomes polarized even though the ensemble of emitting dipoles itself is entirely random. Furthermore, we investigate the effect of the pump polarization and perform numerical simulations to model the polarization state of the emitted light from our bull's eye antenna.