Optical cavities and quantum emitters

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INTRODUCTION

1.1. QUANTUM COMMUNICATION AND QUANTUM MEASUREMENTS

A quantum internet that provides fundamentally secure communications is a long-standing promise from the field of quantum optics [1, 2]. The 2022 Nobel Prize was even awarded in this field. In recent years, researchers have made significant steps towards larger and more efficient networks [3, 4] and have shown how larger distances can be covered with well-functioning quantum repeaters [5]. Furthermore, the field is moving towards the commercial market, signaled by the many start-up companies that are emerging [6, 7].

The reason why there is still no widely used quantum internet is potentially because the ideal quantum emitter is yet to be found. Many research groups have found emitters that fulfill some of the requirements; easy (on-chip) use [8, 9], high brightness and single-photon purity [10, 11], good initialization and manipulation techniques [12], and emission at telecom wavelength [13]. The search for emitters with all of these criteria is still ongoing.

Quantum measurements using photonics is another promising field. Bose-Einstein condensates have resulted in incredibly narrow-linewidth lasers and atomic clocks, which are used to study fundamental fields on physics, like gravitational wave detection [14], dark matter detection [15] and quantum gravity [16]. Other quantum sensors are optically detectable spins in NV centers. These are useful tools for measuring local magnetic fields and are used to detect spin-waves in condensed matter [17]. A promising new field is the observation of spin-defects in van der Waals materials [18], which can probe such spin waves in closer proximity.

1.2. QUANTUM EMITTERS IN AN OPTICAL RESONATOR

A good quantum source of photons requires (i) a reliable single-photon emitter and (ii) an optical resonator to direct the emission and increase the light-matter interaction. By reciprocity, an ideal quantum emitter absorbs (and re-emits) every (strongly-focused) single-photon. For an ideal emitter, the cross-section is of the order of the wavelength λ, $\sigma_{\text{abs}}^{\text{ideal}} = 3\lambda^2/(2\pi)$ [19]. In most cases however, the cross-section is much smaller than this ideal case, for example, for NV centers, $\sigma \sim 10^{-7}\sigma_{\text{abs}}^{\text{ideal}}$ [20] and for rare-earth ions, $\sigma \sim 10^{-12}\sigma_{\text{abs}}^{\text{ideal}}$ [21]. Therefore, in free space, only a portion of the incoming light is absorbed by the emitter, as is shown in figure 1.1a, and demonstrated in experiments [22].

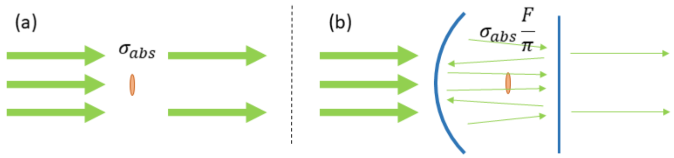

Figure 1.1: Schematic picture of the absorption of an emitter. (a) Most emitters absorb only a fraction of the incoming light beam. (b) Inside the cavity, the light reflects multiple times, such that all light can in principle be absorbed.

An optical resonator, or cavity, can enhance the absorption cross-section, as Fig. 1.1b illustrates. There exist many types of resonators, using dielectric materials [23], (sub-wavelength) antennas [24], or hybrid structures [25]. In this thesis, we focus on the purely dielectric structure of a Fabry-Perot cavity, where the light is reflected many times between the two mirrors. Typical mirror reflectivities are $|r|^2 > 99$ % which leads to a high cavity finesse $F = \pi|r|/(1-|r|^2)$. The number of roundtrips inside the cavity before the photon is lost is F/π. The finesse can also be expressed as $F = \frac{\omega_{\text{fsr}}}{\Delta\omega}$ with free spectral range ω_{fsr} and cavity linewidth $\Delta\omega$. A high finesse cavity strongly increases the effective cross-section to $\sigma^{\text{eff}}_{\text{abs}} = \frac{F}{\pi}\sigma_{\text{abs}}$ [19].

If a cavity is used, the incoming beam is not perfectly focused anymore but depends on the geometry of the cavity. In the case of Fabry-Perot cavities, the mode area of the incoming beam is $A = \pi w_0^2 = \lambda\sqrt{L(R-L)}$ with mode waist w_0, cavity length L and radius of curvature R. The enhancement of the cavity is given by the Purcell factor [26],

$$F_p^{\text{max}} = \frac{\sigma^{\text{ideal}}_{\text{abs}}}{A}\frac{F}{\pi} = \frac{3\lambda^2}{2\pi^2}\frac{F}{A} = \frac{3\lambda^3}{4\pi^2}\frac{Q}{V} \tag{1.1}$$

where Q and V are the cavity's quality factor and mode volume. The quality factor is related to the finesse through $Q = \frac{2L}{\lambda}F$.

Note that the Purcell enhancement can be smaller due to a mismatch of the emitter dipole $\vec{\mu}$ and electric field orientation $\vec{E}$. Furthermore, when the emitter is not placed in the antinode of the cavity mode, the Purcell factor decreases [27], which can be expressed by the factor $\sin^2(kL)$, where $k = \frac{2\pi}{\lambda}$. Last, the emitter frequency ω_e and the cavity frequency ω_c can be detuned, which also reduces the Purcell enhancement. These effects result in a smaller Purcell factor

$$F_p = F_p^{\text{max}} \frac{\mu \cdot E}{|\mu||E|} \sin^2(kL) \frac{\delta\omega^2}{4(\omega_e - \omega_c)^2 + \delta\omega^2} \tag{1.2}$$

The ideal emitter remains coherent for much longer times than photons stay in the cavity. This is not always the case, and in fact, many emitters have quality factors $Q_{\text{emitter}} < Q_{\text{cavity}}$. The Purcell factor in this case is

$$F_p^{\text{eff}} = \frac{3\lambda^3}{4\pi^2}\frac{1}{V}\left(\frac{1}{Q_{\text{emitter}}} + \frac{1}{Q_{\text{cavity}}}\right)^{-1}. \tag{1.3}$$

The equation of the Purcell enhancement shows that in some cases the cavity enhancement is limited by the spectral linewidth of the emitter. In such cases, it is beneficial to reduce the radius of curvature and cavity length as much as possible to obtain a good Purcell factor, as was shown in Ref. [28].

1.3. OPEN MICROCAVITIES

Open microcavities with small radii of curvature are interesting objects to study by themselves. Besides achieving very high Purcell enhancements with quantum emitters [27, 29], empty microcavities with an XY-tuned flat mirror can be used as accurate scanning probes [30–32]. A large part of this thesis studies optical microcavities without quantum emitters. We, among others, observe cavity resonances and modes that cannot be explained by the standard theory for optical cavities.

Figure 1.2 shows a plot of finesse and radius of curvature of microcavities from several research groups. The upper-right corner, where $F\lambda/R > 10-100$, shows the region where a complex mode spectrum can be observed that can not be explained by standard theory of optical cavities [33]. The number of reported cavities is far greater than shown in the figure, but the selected papers already show a variety of fabrication methods and cavity types. The figure shows that our microcavity is not unique. Hence, other research groups must also have seen the results that we have reported. The fact that most of these groups do not report the origin of these effects shows that the complexities in microcavities have not been fully understood yet. Figure 1.2 also shows a blue-shaded area, where $R \lesssim 10\lambda$. In this region, the nonparaxial effects dominate over typical mirror-shape effects, such as astigmatism. This means that even a microcavity with a perfectly spherical mirror shows mode mixing and a so-called fine structure. The figure of merit $F\lambda/R$ to observe (non-paraxial) mode spectra scales in almost the same way as the Purcell factor. Microcavities with a better Purcell factor will also show more nonparaxial effects.

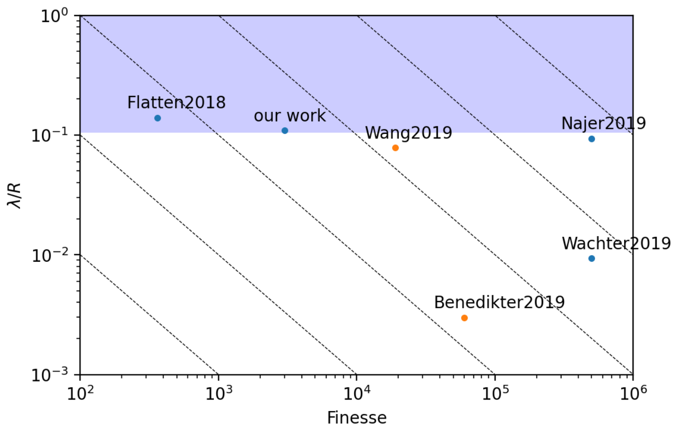

Figure 1.2: A (non-exhaustive) selection of literature reports on open microcavities in terms of their cavity finesse F and normalized inverse mirror radius λ/R. The dashed lines indicate constant values of $F\lambda/R$, where a mode spectrum is generally observable if $F\lambda/R > 10-100$. The blue shaded area, where $\lambda/R > 0.1$, shows where the nonparaxial effects generally dominate over mirror-shape effects, i.e. where the mode structure becomes a fine structure. Points in the figure are taken from: Flatten2018 [28], Wang2019 [27], Benedikter2019 [32], Najer2019 [29], Wachter2019 [34].

1.4. SINGLE-PHOTON EMITTERS

1. An optical cavity also requires a good single-photon emitter. Equation (1.3) shows that the emitter should have a quality factor of the same order as the optical cavity. Next, its

surface should be flat, such that scattering losses do not reduce the finesse too much. Also, the possibility of room-temperature operation increases its usefulness in future applications. Last, a spin ground state with radio-frequency control, like in nitrogen-vacancy (NV) centers, would allow easy initialization and control of the emitter.

One type of emitter that shows promising results towards all the criteria above is hexagonal Boron Nitride [35]. This van der Waals material, which consists of Boron and Nitride atoms in a hexagonal lattice, forms a good insulator with a bandgap of ~ 5 eV. The defects that we have studied are generally assumed to be carbon-vacancies [35, 36]. This is however still debated [37] and only careful experimental work can help this discussion forward [38]. Other similarities of such emitters with the NV centers are their room-temperature operation [39] and their sensitivity to optically detected magnetic resonance (ODMR) which suggests a spin ground state [18, 40]. A 2D material like hBN is especially suited for cavity purposes because its layers are atomically flat. Diamond requires complicated etching techniques making it challenging to maintain a high cavity finesse [41, 42].

Despite all these advantages, a challenge in the use of hBN is low observed count rates. This might be due to a relatively large non-radiative decay rate. We also find that certain emitters show shelving in dark states. The count rates can be enhanced when the emitters are coupled to the microcavity. However, the zero-phonon-lines that we observe at room temperature are relatively broad, $\Delta\lambda_{ZPL} \approx 10$ nm, which is much broader than the cavity linewidth $\Delta\lambda_{ZPL} < 0.1$ nm. We expect this to decrease when cooled to cryogenic temperatures [43]. A cryogenic optical cavity brings about its own challenges with stability and mode-matching. This is left as an outlook for future research.

1.5. OUTLINE OF THIS THESIS

Chapters 2-6 of this thesis describe the formation and characterization of optical modes in cavities. Chapters 2-5 show measurements on an empty cavity with external lasers as the light source, whereas Chapter 6 uses intracavity emitters as an internal light source. Chapter 7 shows confocal microscope measurements on quantum emitters in hBN.

- Chapter 2 analyzes the reflection properties of the Distributed Bragg Reflectors (DBRs) that are typically used in microcavities. It shows that the penetration of the optical field in these mirrors is best described by introducing three penetration depths, where the experiments determine penetration depth should be used.

- Chapter 3 describes the coupling of the fundamental mode with transverse modes. Far-field images show strong deviations from the theoretical description of the uncoupled fundamental mode over a large range of cavity lengths. Additional AFM measurements show that the mode-coupling originates from a shape deviation at the center of the cavity. The measurement of far-field images proves to be a sensitive technique for measuring mode-coupling.

- Chapter 4 gives a theoretical description of a so-called fine structure in microcavities, caused by nonparaxial effects. These nonparaxial effects become important for high-finesse ($F > 1000$) cavities with small radii of curvature, typically $\lambda/R > 0.1$. The theory makes predictions of the mode spectrum for ideal mirrors, but also shows how other deformations influence the mode spectrum.

- Chapter 5 shows the observed fine structure in close-to-perfect microcavities. The mode-structure and (polarization-resolved) far-field images are compared to the theoretical predictions, and show a good match.
- Chapter 6 describes measurements of all of the effects mentioned in Chapters 2-5, but now using intracavity emitters as a probe. This brings new perspectives on the concepts of penetration depth and mode-coupling that are important when cavity emission spectra are probed, rather than length spectra.
- Chapter 7 shows measurements of multiple defects in hBN. Several aspects were characterized, such as their spectrum, saturation behavior, and dipole orientation. A Hanbury-Brown and Twiss measurement reveals interesting population dynamics, for which two models with rate equations are proposed.