Hybrid Resonators for Light Trapping and Emission Control
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Summary

Light is at the heart of many technologies in our modern society. It can visualize tiny cells or entire galaxies, measure sub-nanometer displacements or individual viruses in blood, imprint complex patterns on a microchip or transport enormous amounts of information. For these and other applications, control over the flow of light and its interaction strength with matter is pivotal. Optical resonators, which compress and store light for a finite time, have been instrumental in providing such control. The current state of the art for these resonators consists of optical cavities, which store light for many thousands or millions of oscillations, and plasmonic antennas, which instead squeeze light to sizes much below the wavelength. While these have shown impressive results, both approaches suffer from intrinsic drawbacks that limit large-scale implementations, particularly the realization of a network for optical quantum information processing. This thesis explores how hybrid resonances can overcome these limitations and provide new methods to guide and store light. Such resonances combine two or more resonances in a non-trivial manner, such that the hybrid resonance is more than simply the sum of the individual resonances. In particular, we study two types of hybrid resonances — those arising in a coupled cavity-antenna system (Chapters 2 to 8), and so-called 'bound states in the continuum' (Chapter 9).

Chapter 1 introduces the concept of light-matter interaction and summarizes the current best efforts at controlling this interaction through optical cavities, plasmonic antennas or combinations thereof. We then continue with a theoretical study of hybrid cavity-antenna systems. In Chapter 2, we develop a simple and intuitive model, based on coupled harmonic oscillators, to describe the interaction between a plasmonic antenna and a cavity resonance. Despite its simplicity, the model captures the essential physics of this interaction, and holds for any cavity or antenna geometry. It connects in one framework various properties of such systems, including mode hybridization, cavity perturbation, modifications of the local density of states (LDOS) and photon collection efficiency. This chapter lays the foundations for analyses performed in several subsequent chapters.

Chapter 3 employs the coupled-oscillator model to study theoretically the LDOS in cavity-antenna hybrids. We demonstrate that hybrids can support larger LDOS than their bare constituents. The conditions for such symbiotic behaviour, however, are far from trivial. We find that, contrary to intuition, operating near antenna resonance is detrimental to LDOS due to destructive interference between cavity and antenna. In contrast, at frequencies red-detuned from the antenna resonance, constructive interference can lead to very large LDOS, even breaking a fundamental limit governing the LDOS for a single antenna. Moreover, we show how hybrid systems can be designed to support quality factors Q and mode volumes V anywhere in between those

of the cavity and of the antenna. Also, photon collection efficiency can be high, despite plasmonic losses. These properties make them very attractive candidates for single-photon sources, which require efficient operation, large LDOS and bandwidths matching those of realistic emitters.

Chapter 4 applies concepts from electrical engineering to hybrid cavity-antenna systems by developing a circuit analogy for these hybrids. Since this is based on an analogous circuit for a nano-antenna, we first discuss two different versions of such a circuit from literature and show that the two are equivalent. We then find the circuit for a hybrid system and show how a cavity can help an antenna reach an upper limit on radiation, set by the maximum power transfer theorem. This limit compliments the well-known unitary limit on antenna radiation, and we elucidate the interplay between these limits for lossy antennas. Our results show that cavities act as conjugate-matching networks at optical frequencies, matching the antenna to its radiation load. Such networks allow maximum power transfer from a lossy generator to a load and are common in radio-frequency network engineering, yet they have remained elusive at optical frequencies.

Having established theoretically that hybrid systems are a promising platform for strong light-matter interactions, and having found design rules to benefit optimally from these interactions, Chapter 5 then takes us to the experimental work. This chapter describes the fabrication of cavity-antenna systems, loaded with fluorescent quantum dots. Hybrid systems composed of whispering-gallery-mode microdisk cavities and aluminium antennas are fabricated with high precision and repeatability using a two-step lithography process. A study of LDOS effects requires fluorescent emitters, positioned accurately in the hybrid system. We present a novel method to deterministically position fluorescent colloidal quantum dots in such systems, which uses a PMMA resist mask and a linker molecule to covalently bind the dots to the hybrid structures. This allows highly selective placement of quantum dots with lithographic resolution.

The hybrid systems fabricated as described in Chapter 5 are studied experimentally in Chapter 6. A crucial property of cavity-antenna hybrids is the ability to tune the bandwidth over a large range, which enables coupling to realistic emitters. Here, we experimentally observe more than two orders of magnitude linewidth tuning in hybrid systems, simply by changing antenna length. For this, we measure antenna-induced linewidth broadening and shifts for antennas and disks of various sizes, using a combination of tapered-fiber spectroscopy and free-space microscopy. We show that our results can be explained using cavity perturbation theory, and observe a deviation from this theory at antenna sizes for which the dipole approximation breaks down.

In Chapter 7 we put the theoretical predictions from Chapter 3 — which state that LDOS can be strongly boosted in a hybrid system — to the test. Fluorescence spectra, measured on the hybrid systems loaded with quantum dots, reveal Fano-type asymmetric resonances. These arise due to quantum dot emission into the hybrid modes. Our results demonstrate that LDOS is strongly boosted at these modes, up to 14 times higher than the LDOS provided by the antenna alone. While LDOS is commonly measured through a change of the fluorescent decay rate of the emitter, we argue that, for a broadband emitter coupled to a narrowband LDOS resonance, *relative* LDOS can be obtained from the emission spectrum. Our analysis is supported by an excellent agreement between theory and experiment in the linewidth, -shape and fluorescence boosts. Additionally, fluorescence decay rate measurements show a strong increase of decay rate, which we attribute mainly to the antenna. Combined, these results demonstrate that large LDOS enhancements are possible in hybrid systems.

Rich new physics arises when a cavity is coupled not just to a single antenna, but instead to a lattice of antennas. In Chapter 8 we experimentally study such an antenna lattice, coupled to an ultra-high-Q microtoroid cavity. Through an intricate extinction measurement performed in reflection, we find that back-action by the cavity on the antennas causes a strong suppression of the antenna polarizability. This matches the theoretical prediction in Chapter 3 for a single antenna, which stated that an antenna is spoiled by a cavity if both are on resonance. There are, however, important differences between a single scatterer and a lattice. We show that in a lattice, antenna-cavity coupling can be modified by changing the angle of incidence, because this coupling requires momentum-matching between the Bloch waves in the lattice and the cavity mode.

The final Chapter 9 of this thesis studies hybrid resonances of a different type. Bound states in the continuum (BICs) are modes found, for example, inside a photonic crystal slab, which are normally leaky yet become perfectly bound at one particular wavelength. Such states were observed experimentally only recently in optics and are puzzling from a fundamental point of view, as well as interesting for applications due to their (theoretically) infinite photon lifetime. We experimentally demonstrate that such a state coincides with a polarization vortex in momentum-space. To do so, we develop a new ellipsometry method to quickly measure polarization-resolved reflection at many angles and wavelengths. The existence of the vortex implies that the BIC is topologically protected, such that it is robust against small variations in geometry. Moreover, we demonstrate that a hybridized resonance underlies the perfect confinement of this mode. An electric and a magnetic dipole resonance inside the crystal unit cell couple through interference in the far-field, leading to a perfectly bound state when complete destructive interference between the two is achieved.