Quantum dots in microcavities: from single spins to engineered states of light

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1 Introduction

1.1 Single photons

Photons, the elementary quantum particles of light, are truly fascinating objects. Some studies even suggest that the human eye can register them individually, despite typically being processed in much larger quantities [1]. The availability of sources of entangled photons has made photons the most popular platform to demonstrate quantum phenomena such as quantum superposition, quantum non-locality [2], and quantum teleportation [3]. Since then, scientists fascinated by quantum theories have designed a variety of applications based on these properties. For example, many photons in linear optical networks have recently achieved an important demonstration of the "quantum advantage" [4]. Also, for universal quantum computation, photons have potential, either in the form of many single photons [5–7] or using multi-photon entangled states such as cluster states [8–11] as a resource. In the one-way quantum computing scheme [8], challenges from probabilistic or deterministic two-photon gates are shifted to the engineering of large-scale cluster states [12]. The weak interaction with the environment protects photons from decoherence, but this also limits their interaction with matter or with other photons, therefore it is in fact hard to generate both single photons and cluster states in the first place.

In this thesis, we follow one of the roadmaps of single and entangled photon generation – using a III-V self-assembled InGaAs quantum dot (QD) embedded in an optical microcavity. Electrons and holes in the QD have discrete energy levels similar to atoms. Under resonant excitation, the QD can be considered a two-level system and transitions between the states happen by the absorption or emission of a photon. Like for atoms, this interaction is in homogeneous materials not efficient, and therefore, we embed the quantum dot in a Fabry-Perot cavity formed by two highly reflective mirrors. This leads to an increased interaction of light with the QD, and allows for deterministic photon-matter interaction. Furthermore, inserting the QD into a cavity leads to acceleration of the emission into the particular cavity mode, known as the Purcell effect, with two important effects on single-photon generation. First, the single-photon collection benefits from this effect because the QD preferentially emits into the selected cavity mode. Second, the Purcell effect shortens the recombination time of the QD electron-hole pair, leading to higher photon indistinguishability due to a reduced influence of dephasing. Together with the fact that a two-level system can only emit one photon at a time, the QD-cavity system is a near-ideal source of single photons and allows for photon generation on demand [13–15].

Tremendous progress has been made in the last decade with QD-cavity–based single-photon sources [16], where scientists carefully optimized every single device parameter [17] and brought these sources close to perfection. The best single-photon sources now show excellent single-photon purity (97.9 %), indistinguishability (97.5 %), and brightness (57 %) [15]. The possibility to collect photons efficiently with an optical fiber directly attached to the top of the single-photon-source semiconductor chip [18] and commercialization thereof [19] makes these sources also very appealing for wide distribution and application in a variety of quantum technologies. High-quality true single-photon sources would be a crucial advantage for optical quantum computing approaches.

For applications in quantum networks, key challenges are the indistinguishability of photons produced by separate single-photon sources and the photon wavelength, which should be compatible with telecom technologies. Because QD properties, including emission wavelength, can be tailored by the nanostructure composition, scientists are positive about a bright telecom single-photon near-term future. Already now, the first telecom single-photon sources have been reported, either based on InP technology [20, 21] or frequency conversion [22]. Yet, the preparation of two (or more) identical single-photon emitters necessary for large quantum networks is still an open problem, mostly due to limited control of the atomic-scale composition of individual QDs during their self-assembly [23]. Only recently, advanced QD growth techniques such as droplet etching epitaxy [24, 25] have allowed demonstration of two-photon interference with near-unity visibility (93.0 %) using photons from two completely separate quantum dots [26].

1.2 From single to entangled photons

Having access to QD-cavity-based sources of high-quality and indistinguishable single photons allows us to manipulate them and synthesize never-produced complex quantum states of light. This simply requires an optical beam splitter: If two indistinguishable photons arrive simultaneously at the two inputs of a beam splitter, the photons will "bunch" and exit the beam splitter always together through one exit port. This result is very different from the classical situation, where there is always a finite probability that photons leave the beam splitter separately. This photon bunching is a purely quantum effect called Hong-Ou-Mandel (HOM) interference [27] and allows, for example, a direct comparison of two states of light. This state comparison is used to experimentally characterize the photon indistinguishability of single-photon emitters [28] or for guaranteeing security in quantum network protocols [29].

In quantum optics, by only using beam splitters and phase shifters, one can construct arbitrary unitary operations on quantum states of light for quantum information processing [30, 31], where the HOM effect plays an essential role. On the long way towards the processing, there is a simpler interesting application of repeated HOM interference - boson sampling [32], where several indistinguishable photons in Fock states are combined in a large free-space or integrated-optics interferometer [4, 33–35]. This interferometer or photonic circuit implements an unitary operation from the inputs to its outputs. The outcome of this circuit is computationally hard to calculate on classical computers [36], making boson sampling useful to demonstrate a "quantum advantage". A few proof-of-principle experiments with photons have been reported, including realizations with de-multiplexed single photons from a QD-cavity system [34, 35]. The largest photonic interferometer to date with true single photons was implemented in free space with 60 input and output ports and 20 single photons prepared by de-multiplexing of a QD-cavity single-photon source [37]. Despite the enormous size of the Hilbert space accessible in the experiment (3.7×10^{14}), corresponding to 48 qubits, the interferometer was built only for a specific unitary operation. Currently, scientists are developing large, scalable, and programmable photonic chips (or processors) [38], enabling the implementation of arbitrary linear optical transformations necessary for universal photonic quantum computing.

Moreover, having these processors, we can engineer complicated photonic states, including multi-photon entangled states such as cluster states [39], for other quantum applications. Sometimes, for a specific application, the interferometer can be strongly simplified.

For example, using time-bin photon encoding, boson sampling can be performed with a single beam splitter, if photons from one exit mode are directed back to the same beam splitter, forming an optical delay loop [40, 41]. Here, the delayed loop-photons interfere with single photons from the source and develop complex photon statistics [41]. A similar optical delay loop was used to entangle sequentially emitted single photons from a QD-cavity device into a linear cluster state [42]. In general, entanglement of many photons using linear optical interferometers is often not fully deterministic, this is where the QD again provides solutions as we discuss now.

1.3 The QD as a spin-photon interface

Another remarkable property of quantum dots is to trap a charge carrier. This confined extra electron or hole provides a spin to the originally empty ground state. This spin can be used as a quantum memory with information encoded in the spin orientation [43]. The extra charge changes the QD optical selection rules connecting the photon polarization to the spin state, therefore the charge level of the QD can be confirmed by polarization analysis of the single photons emitted by the QD [44].

The singly-charged QD has two ground states of opposite spin. These states are naturally energy degenerate but can be split by the Zeeman effect in an external magnetic field. This splitting enables spin initialization [45,46] and read-out [47] with optical fields, therefore entangling photons with the single spin. Due to the recent impressive improvement of the spin coherence time up to $T_2 = 0.113\,\mathrm{ms}$ [48], the QD spin can be entangled with many photons during the spin coherence time. This, in combination with entanglement swapping [49] using the HOM effect at a beam splitter, is an essential tool to establish a quantum link between distant places. Integrating the charged QD into an optical cavity to accelerate the spin-photon entangling rate would further increase the remote-entanglement generation rate, but cavity-QD devices with both high Purcell-accelerated emission and extra-long spin coherence still have to be optimized.

Moreover, spin-photon entanglement generation in combination with spin precession in a weak magnetic field also allows for the deterministic generation of linear cluster states [50]. Here, each emitted photon is entangled with the same spin, and the spin can be "traced out" in the end to obtain a purely photonic entangled state. Despite being deterministic in principle, this scheme has been experimentally limited by reduced photon collection efficiency to less than 1% due to the absence of a cavity [51, 52]. Only recently, using a charged exciton in a Fabry-Perot cavity, the group of Pascale Senellart achieved three-particle entanglement with fidelities up to 63% [53].

1.4 Resonant laser spectroscopy

Any successful quantum protocol starts with qubit initialization, often carried out by excitation with a resonant laser. In singly-charged QDs, successful spin initialization can be detected by observation of the QD resonant fluorescence because of the optical selection rules. For this, the weak QD emission has to be separated from the stronger excitation laser light. This laser filtering is typically done in the polarization degree of freedom using cross-polarization where the excitation and detection polarizations are orthogonal, and cross-polarization extinction ratios up to 10^8 [54] have been achieved. Surprisingly, these ratios are more than one order of magnitude higher than expected from the specification of the polarizers. The physical origin of this has been an open question until recently [55], and turns out to be a combination of residual ellipticities and spin-orbit coupling of light. Since QD-cavity systems require cryogenic temperatures, it is essential to also achieve high extinction ratios in a cryostat - which is complicated by several issues, from the cavity birefringence to strained cryostat windows.

1.5 Thesis outline

In Chapter 2, we introduce and characterize our cavity device containing singly-charged QDs. Further, we present a resonant spectroscopy toolbox used for QD spin-state identification and characterization.

In Chapter 3, we experimentally study cross-polarization extinction in several optical setups, including firstly in a cryogenic confocal microscope. We find that using optical reflection with single-mode filtering detection leads to potential cross-polarization extinction improvement above the limit given by bare polarizers. This improvement is associated with Fresnel-reflection-induced effects, including the spin-orbit coupling of light, allowing for small pre-compensation of the residual ellipticity of linear polarizers. We identify this effect in confocal microscopy and use it to demonstrate improvement in single-photon emission from a singly charged quantum dot in a birefringent optical microcavity.

In Chapter 4, we develop a cryogenic permanent (Voigt geometry) magnet assembly and use it to split the electron spin states of a charged QD resonant with a micropillar cavity. Then, using two-color resonant spectroscopy, we demonstrate spin population manipulation by controlling the relative power of the excitation lasers, and we observe an increase in single-photon emission.

Adding a single photon on a highly reflective beam splitter to a classical optical field is a known but experimentally challenging technique to prepare complex multi-photon quantum states. In Chapter 5, we theoretically study this scheme for the generation of displaced Fock states with various single-photon sources. By analytical theory, we show that photon-correlations of the produced states exhibit a global maximum as a function of the strength of the single-photon state. Further, we discuss experimental realization and the use of photon correlations to optimize the mode-matching between single-photon and coherent states.

In Chapter 6, we use our QD-cavity device as a single-photon source, and by repeated quantum interference we build from the single-photon stream complex photon-number superposition states with tunable photon statistics, including approximately coherent states of light. The engineered artificial coherent states are more complex than the conventional coherent states, containing quantum entanglement of photons, making them a promising resource of multi-photon entanglement.