Chapter 1

Introduction

Optical microcavities are used to confine light both spatially and temporally. The spatial confinement is typically described by the mode volume (V), whereas the temporal confinement is described by the quality factor (Q), which is the photon storage time, normalized with respect to the frequency of oscillation. Surface-tension-induced-microcavities, such as micro droplets or microspheres, are dielectric cavities which are created by surface tension and which exhibit a near atomic scale surface finish. Light within the sphere is confined by continuous total internal reflection near the cavity perimeter, and the modes have therefore been called "whispering-gallery" modes. Of all geometries studied for confining light, surface-tension induced silica microspheres have attained the highest optical quality-factors (Q) to date of nearly 10 billion [1], and are of interest for a variety of studies ranging from fundamental physics such as cavity Quantum Electrodynamics [2][3][4] to more applied areas such as low threshold and narrow line-width lasers [5][6][7], as well as high-sensitivity transducers for biochemical sensing[8]. The small mode volume and long photon storage time can also be used for nonlinear optical studies, as strong resonant buildup of energy in micro-scale volumes significantly reduces the threshold for nonlinear optical effects to occur. This was recognized in the pioneering work of Chang [9][10] and Campillo [11][12][13]who observed and studied a variety of nonlinear optical effects in ultra-high-Q liquid micro-droplets. Their work used free-space illumination to optically pump the micro-droplets and thereby induce Raman oscillation [10][11][13], cascaded Raman scattering [10] and Brillouin scattering[14]. However, due to their transient nature, liquid micro-droplets have remained a mere laboratory tool, and the pump threshold for nonlinear effects have remained high, despite ultra-high Q, due to the low efficiency of free-space excitation. Furthermore, despite a wealth of reported nonlinear optical microcavity effects, *parametric* oscillation has not been observed so far.

Silica microspheres provide a far more stable and robust microcavity in comparison with liquid micro-droplets. However, despite numerous studies on these devices over the past decade [15][16][17][18][19][20][21] [22][23]the observation of nonlinear phenomena (beyond thermal effects) in these devices, had been limited to one report on Kerr-induced wavelength shifts at low temperatures [18].

1.1 Thesis outline

In this thesis the stimulated and parametric nonlinear optical processes in ultra-high-Q silica microcavities are investigated and analyzed for the first time. To pump the optical whispering-gallery modes of the silica microcavities efficiently, tapered optical fibers were used[24][25]. It is demonstrated that the excitation using tapered optical fibers can be highly efficient [26], and can allow to couple to silica microcavities with negligible parasitic (junction induced) loss. Ultra-high-Q microcavities naturally transit into a regime, where surface scattering centers can render the degenerate clockwise and counterclockwise mode strongly coupled, giving rise to the regime of strong modal coupling[27]. It is shown that in this regime the tapered-optical fiber coupling properties are significantly altered. The whispering-gallery modes appear significantly split, and behave as a frequency selective mirror. It is shown, that even in the presence of modal coupling, high circulating power within the cavity can be achieved. and allowing to exceed the threshold for all common nonlinear optical effects of silica. Stimulated Raman scattering, the interaction of light with optical phonons of silica, is observed in fiber-coupled silica microspheres and the measured threshold for nonlinear oscillation are lower than for any other nonlinear optical oscillator reported to date. In particular, a silica micro-sphere Raman lasers with ultra-low threshold levels of only 62 μ-Watts [28] is demonstrated in this thesis. Compared to micro-droplets these devices allow stable and long term observation of nonlinear optical effects in microcavities. Cascaded Raman lasing in these devices of up to 5 orders has also been observed [29][5] and the lasing properties analyzed theoretically and experimentally. The tapered optical fiber in these experiments functions to both pump WGMs as well as to extract the nonlinear Raman fields. In addition, the tapered-fiber coupling junction is highly ideal[26], making it possible to strongly over-couple ultra-high-Q cavities with negligible junction loss. This feature allows for the observation of very high *internal* differential photon conversion efficiencies approaching unity[5].

Whereas microspheres are both compact and efficient nonlinear oscillators, their fabrication properties lack the control and parallelism typical of micro-fabrication techniques. In this thesis the optical properties of toroid microcavities on-a-chip [30] are analyzed, and ultra-high-Q modes (UHQ) demonstrated. The measured Q-factors in this thesis constitute an improvement in Q-factor of nearly 4 orders of magnitude compared to other chip-based microcavities. UHQ toroids have several advantages over spheres including being wafer-scale devices that can be fabricated in parallel as dense arrays or integrated with electronics or complementary optical functionality. The use of toroid microcavities as nonlinear oscillators is investigated, and the first Raman oscillator on a chip is demonstrated[8]. The reduced mode volume of toroid microcavities, allow to observe stimulated Raman scattering at effectively lower threshold than in microspheres. In addition the strongly reduced azimuthal degree of freedom, allowed to obtain single mode Raman lasing, over a large range of pump powers. This constitutes a significant advantage over micro-spheres and micro-droplets. Furthermore, the effect of the toroid geometry on the nonlinear optical effects is studied and is found to profoundly alter the nonlinear optical processes in the microcavity. Specifically, a reduction of the toroid cross sectional diameter, allowed to induce a shift from stimulated Raman to Parametric oscillation regime. This allowed to observe Kerr nonlinearity optical parametric oscillation in a microcavity for the first time. Optical parametric is observed at ultra-low threshold and high efficiency, and the generated signal-idler "twin beam" show near unity signal-to-idler ratio[9].

1.2 Chapter overview and collaborative work

The results of the author presented in this thesis were to a large extent performed in collaborative work with his colleagues. In what follows the results of the individual chapters are given and the relative contributions indicated.

Chapter 2 is a self contained introduction to spherical dielectric resonators (silica microspheres). The resonant characteristics of microspheres, such as their field distribution, mode volume and radiation loss are discussed and serve as an introduction to the terminology which is used throughout this thesis.

Chapter 3 describes the experimental infrastructure the author has implemented jointly with his colleague Sean Spillane to continue work on microspheres resonators, that had initially be been started by graduate student Ming Cai. The experimental setup for fabrication of tapered optical fibers, as well as measurements the author performed on silica microspheres are described. The up-conversion pictures in chapter 1 were obtained from microspheres which were implanted with erbium ions, a task Jeroen Kalkman from the group of Albert Polman at AMOLF accomplished.

Chapter 4 investigates the influence of strong mode splitting (which is commonly observed, due to the sensitive nature of the ultra-high-Q microcavities to surface defects) on the coupling properties of tapered optical fiber. In particular, the author has observed and described the coupling properties in the regime of strong modal coupling, in which the resonator mimics a frequency selective reflector. This chapter has appeared in *Optics Letters [27].* In addition these measurements were carried out in the presence of negligible parasitic loss, a property which has been further investigated by his colleague Sean Spillane and which has appeared in *Physical Review Letters*[31].

Chapter 5 describes the observation of ultra-low threshold stimulated Raman lasing in taper-fiber coupled silica microspheres, which the author studied and explored in collaboration with his colleague Sean Spillane. Threshold values which are more than 3 orders of magnitude lower than in previous work have been obtained. This chapter has been published in *Nature[31]*.

Chapter 6 presents a theoretical and experimental analysis of stimulated and cascaded Raman scattering in taper coupled micro-spheres resonators, and complements the results of chapter 4. The author investigated the properties of cascaded Raman scattering theoretically, which Bumki Min verified experimentally. This chapter has been published in *Optics Letters [32]* and *IEEE Journal of Quantum Electronics* [5].

Chapter 7, describes the optical properties of the whispering-gallery type modes of toroid microcavities. The author describes a cavity ring-down measurement setup he implemented to obtain measurements of Q-factor. The method allowed to accurately measure photon lifetimes, at high circulating cavity intensity. Using this technique, the author spend one summer continuing measurement his colleague Sean Spillane and Deniz Armani had started, in trying to observe ultra high-Q factors in toroidal microcavities on a chip. The author was finally was successful in the summer of 2002, in demonstrating ultra-high-Q modes in a toroid microcavity on a chip. The samples for these measurements have been made by the author's colleague Deniz Armani. The measurements in this chapter have been published in *Nature[33].* In addition the author investigated the extend to which the micro-torpid geometry could be reduced, and demonstrated a Q/V ratio of more than $10^6\,(\lambda/n)^{-3}$.

In **Chapter 8** the author investigates the optical modes of disk microcavities. Disks and toroid microcavities can be excited efficiently using tapered optical fibers. Surprisingly, disk microcavities allow to observed Q-factors as high as 10^7 which is already nearly 3 orders of magnitude higher than in any other reported chip-based whispering-gallery devices (but lower than in toroid microcavities). The effect is attributed due to the wedge shaped cavity boundary which causes modal isolation. This chapter appeared in *Applied Physics Letters[34]*.

Chapter 9 demonstrates the first Raman oscillator on a chip using toroid microcavities. Raman oscillation in toroid microcavities is compared to micro-sphere resonators, and found to exhibit intrinsically more favorable properties, such as single mode emission and lower effective pump threshold. The work heavily relied on microcavities with ultra-high-Q optical modes, which the author fabricated and obtained with very high reproducibility. This work has been published in *Optics Letters[8].*

Chapter 10 presents the first observation the author made of Kerr nonlinearity induced parametric oscillation in a microcavity. The cavity in this regime parametrically converts a pair of pump photons into a frequency down-shifted signal and frequency up-shifted idler photons, and near unity signal-idler ratio is observed. As parametric interaction does not involve coupling to a dissipative reservoir, and the parametric process creates simultaneously signal-idler photon pairs, the emitted light should exhibit non-classical correlations. The numerical simulations in this chapter have been carried out by the author's colleague Sean Spillane. The author submitted this work to *Physical Review Letters[9].*

In **Appendix A** the coupled wave equations for third order nonlinear phenomena are derived. Starting from the coupled wave equations for plane waves, the equations are reformulated for the whispering-gallery modes of a microcavity. The modified coupling coefficients are given, and the definition of the effective mode volume is discussed.

In **Appendix B** the Helmholtz equation is derived for the case of a whispering gallery mode resonator. The optical modes were numerically modeled using a finite element PDE eigenmode and eigenfrequency solver and the numerical results are compared to analytical solutions and good agreement was found. The method in this chapter has been used for the numerical simulations presented in chapter 7,8,9.

In **Appendix C** the fabrication of toroid and disk microcavities is described. The fabrication sequence was originally started by Deniz Armani and Sean Spillane. The author has made the fabrication process more reproducible (in terms of obtaining ultra-high Q factors). A variation of the fabrication technique investigated by the author is also presented, which allows to obtain more geometric control over the toroid geometry parameters.