Correlated photon-pair generation in a liquid-filled microcavity

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Abstract

We report on the realization of a liquid-filled optical microcavity and demonstrate photon-pair generation by spontaneous four-wave mixing. Our source has a spectral brightness of \(45 \pm 7 \text{ mW}^{-2} \text{s}^{-1} \text{ MHz}^{-1}\) and the bandwidth of the emitted photons is \(\sim 300 \text{ MHz}\). We demonstrate tuning of the emission wavelength between 770 and 800 nm. Moreover, by employing a liquid as the nonlinear optical medium completely filling the microcavity, we observe more than a factor \(10^3\) increase of the pair correlation rate per unit pump power and a factor of \(1.7\) improvement in the coincidence/accidental ratio as compared to our previous measurements.

1. Introduction

Nonclassical states of light, such as correlated photon pairs [1, 2] or anti-bunched photons, have contributed significantly to the tests of fundamental quantum mechanics and to interconnect remote quantum systems [3]. One particular challenge when coupling different quantum systems is to match their wavelengths [4, 5] or to bridge wavelength gaps. Different methods from nonlinear optics, such as spontaneous parametric downconversion (SPDC) and spontaneous four-wave mixing (SFMW), have been employed to generate correlated photon pairs. SPDC sources with short crystals [6–11] and SFMW sources, for example in optical fibers [12, 13], employ short-pulse pump lasers propagating through nonlinear optical media. The simplicity of these schemes results from the weak requirements regarding phase matching and leads to correlated photon pair emission in a broad bandwidth of several THz. In contrast, narrow-band sources for photon pairs have often utilized nonlinear media in optical cavities in order to enhance the field strength and control the emission bandwidth. For a recent compilation of parameters see [14–18].

Little attention has generally been devoted to the generation of photon pairs in liquid nonlinear media [19], and no experiments in optical cavities have been reported. In this work, we propose and demonstrate a novel approach of using a liquid-filled optical microcavity to prepare correlated photon pairs with tuneable wavelength separation by SFMW. The four-wave mixing process annihilates two photons from the continuous-wave pump light field at frequency \(\omega_0\) and produces photon pairs at frequencies \(\omega_{\pm} = \omega_0 \pm \omega_{\text{FSR}}\), where \(\omega_{\text{FSR}} = \frac{\pi c}{L}\) denotes the free spectral range of the Fabry–Perot cavity of length \(L\), \(c\) is the speed of light, and \(n\) is an integer. In principle, the usable range of \(n\) is only limited by the bandwidth of the high-reflectivity coating of the cavity mirrors and the transparency of the liquid. The liquid-filled approach of the optical cavity has significant advantages: (1) many liquids exhibit significantly higher nonlinear refractive indices than solids, and (2) there is no additional interface between the nonlinear medium and mirrors, which would affect the cavity performance. The latter is an issue, in particular, for the highly-curved mirror of our microcavity.

The optical Kerr effect in solids has a very fast response time, typically, in the few femtosecond range or below. This has been confirmed by experiments with attosecond laser pulses [20] and rapidly-oscillating optical fields [21]. In liquids, however, the situation is more complex [22, 23]. The optical Kerr effect has a contribution from both the purely electronic degree of freedom and the reorientation dynamics of the molecule if it has an anisotropic polarizability. The latter has a complicated dynamic as it involves intermolecular interactions as well as vibrations and rotations. There has been a long-standing tradition to study the optical Kerr effect in liquids using pump/probe schemes with adjustable time delay and it has been found that the peak Kerr response has
delays with respect to the pump field in the few-ps range and that the instantaneous response is smaller by one or two orders of magnitude as compared to the peak response [24].

2. Experiment

We have constructed a Fabry–Perot microcavity composed of a micromachined and coated endfacet of an optical fiber as one mirror [21, 25–32] and a conventional planar mirror with identical coating as the second mirror (see figure 1). The length of the cavity is \( L = 38.4 \, \mu m \) and the finesse is \( F = \pi/(T + L) = 12,500 \pm 500 \) with a nominal mirror transmission of \( T = 100 \, ppm \) and intracavity losses of \( L \approx 100 \, ppm \) per mirror. The radius of curvature of the fiber mirror is \( R = 200 \, \mu m \), giving rise to a \( 1/e^2 \)-beam radius on the planar mirror of \( w_0 = 3,5 \, \mu m \). This small mode waist enhances the desired nonlinear effects.

We have filled the cavity with the synthetic silicone oil (Tetramethyl–tetraphenyl–trisiloxane) with high optical transparency. The refractive index of the oil leads to an increase of the optical path length of the cavity which has been detected by a change of the free spectral range from \( (3.901 \pm 0.001) \) to \( (2.507 \pm 0.002) \) THz. From this we have determined the refractive index of the oil as \( n = 1.556 \) [33]. However, the absorption coefficient of the oil has not been accurately determined previously. After filling the microcavity with the oil, we have not observed a change of the cavity finesse. Instead, the cavity linewidth decreased from 313 to 200 MHz. Including the effect of the changing mirror reflectivity in presence of the oil, this sets an upper limit on the absorption coefficient of \( \alpha = 5 \, m^{-1} \), which is comparable to pure SiO\(_2\) and indicates that the oil is a very high quality optical medium.

Strong pumping of the cavity with intracavity intensities of up to \( 10^{11} \, W \, m^{-2} \) leads to significant thermal effects. For example, we observe the characteristic bistable cavity line shape, which is a result of the optical path length change caused by absorption-induced heating from the high-intensity pump field inside the oil. To lowest order, it can be modeled by an additional power-dependent detuning in the Lorentzian cavity lineshape [34]

\[
\frac{P_{\text{out}}}{P_{\text{in}}} = \frac{1}{(\Delta - \beta' P_{\text{cav}})^2 + 1}
\]

with the detuning \( \Delta = \nu - \nu_{\text{res}}, \) the natural cavity linewidth \( \delta \nu, \) and the lineshift \( \beta = \beta' P_{\text{cav}}. \) The lineshift broadens the resonance by several tens of the natural linewidth (see figure 2). We measure the lineshift as a function of the transmitted power and observe a linear behavior (see figure 2(c)) [34]. From this we deduce a constant outcoupling efficiency and finesse, even for increasing powers, and hence the intracavity power can be determined from the transmitted power. Moreover, we estimate the temperature increase inside the cavity by connecting the resonance shift to the temperature increase \( \delta T \) inside the cavity via

![Figure 1. Schematic setup of the experiment.](image-url)
with the quality-factor \( Q = 2 \times 10^6 \), the thermo-optic coefficient \( C_{\text{TO}} \) and the refractive index \( n = 1.56 \). The tabulated thermo-optic coefficients of various silicone oils are in the range of \( C_{\text{TO}} \sim 3 \times 10^{-4} \text{ K}^{-1} \) [36], however, the value for our specific oil is not known. Using the average value, we estimate a temperature increase of \( \delta T = 0.2 \text{ K} \). The comparatively low temperature increase results both from the low absorption coefficient and from the fact heat convection in liquids leads to a much faster heat dissipation than heat conduction alone, which would be the mechanism in solids.

In order to minimize variations resulting from thermal effects, we lock the cavity to the pump laser using a Pound–Drever–Hall locking scheme and as a result the residual power fluctuations are below 2%. Using two additional lasers, we measure the frequency difference to the higher frequency \((+\)\) and lower frequency \((-\)\) longitudinal modes for the same order \( n \). We then shift the center frequency of the cavity to the dispersion-compensated point such that both free spectral ranges are equal to within the cavity linewidth. This dispersion compensation plays the role of fulfilling both the phase matching condition and the energy conservation. Generally, we find that different pump power levels affect the dispersion compensation and we perform the compensation individually for each power.

The output of the Fabry–Perot cavity is spectrally dispersed by a home-built grating spectrometer, with a resolution of \( \lambda/\delta\lambda = 6000 \) and the output is recorded with a pair of single photon counters (SPCM). Additional
dielectric line filters protect the SPCM from stray light. Both line filters have a linewidth of 3 nm and transmit light at the respective detection wavelengths while providing a suppression of the pump stray field on the SPCM of at least four orders of magnitude. The measured quantum efficiencies of the two beam paths from the cavity are 9.9% and 7.2% including output coupling from the cavity and photon detection efficiencies. We record the SPCM signals using a time-to-digital converter with timing resolution of 40 ps, much better than the SPCM timing jitter of 350 ps, and subsequently perform a correlation analysis with adjustable bandwidth.

3. Results

In figure 3(a) we show a typical two-photon correlation measurement of order \( n = 2 \), i.e. with photon frequencies of \( \omega_{\Delta -} = 2\pi \times 377.155 \) and \( \omega_{\Delta +} = 2\pi \times 387.155 \) THz for an intracavity power of 0.58 W. The correlation signal shows a coincidence-to-accidental ratio (CAR) of \( 3.4 \pm 0.5 \) and thus clearly signals the nonclassical nature of the emitted photon pairs [37]. The full width at half maximum of the correlation peak is \( 1.06 \pm 0.08 \) ns, which corresponds to a cavity linewidth of \( 328 \pm 19 \) MHz. We attribute the increased width to a higher transmission of the dielectric coating at signal and idler frequencies than at the pump frequency.

In figure 3(b) we show the scaling of the rate of photon pairs versus intracavity power. As expected for a SFWM process, we observe a quadratic dependence on pump power. We compute the expected flux of photon pairs from SFWM from the optical cavity as [38]:

\[
\Gamma = \frac{\omega_{FSR}}{\pi^2} \left[ k \int n_2(z) I(z) \, dz \right]^2.
\]

The quantities \( n_2(z) \) and \( I(z) \) are the nonlinear refractive index and the light intensity, respectively, and \( k = 2\pi n / \lambda_{vac} \) is the wave vector of the light with the refractive index \( n \). The intensity is connected to the intracavity power via \( I = P / \pi w_0^2 \). Note that the quantity \( n_2 \) is proportional to the Kerr constant evaluated at an optical frequency and hence will be lowered as compared to the tabulated dc (\( \omega \to 0 \)) value of \( n_2^d = 44 \times 10^{-20} \text{ m}^2 \text{ W}^{-1} \) [39] by the mechanisms discussed in the introduction. In principle, the integral over the Kerr coupling would even include the nonlinear effects arising from the mirror coating [40, 21], however, they are approximately two orders of magnitude smaller and can be neglected. The experimentally determined rate coefficient with respect to the intracavity power \( \Gamma_{exp} = 1.12 \pm 0.05 \text{ W}^{-2} \text{ s}^{-1} \), see figure 3(b), corresponds to a theoretical coincidence rate in relation to the pump power of \((14,800 \pm 2100) \text{ mW}^{-2} \text{ s}^{-1}\). The resulting spectral brightness is \((45 \pm 7) \text{ mW}^{-2} \text{ s}^{-1} \text{ MHz}^{-1}\). We deduce the nonlinear refractive index \( n_2 = (3.62 \pm 0.31) \times 10^{-20} \text{ m}^2 \text{ W}^{-1} \), which is one order of magnitude lower than the dc-value and comparable to SiO₂.

On both spectrometer channels (+\( n \)) and (−\( n \)) we find a linear dependence of the count rate \( R_{\pm n} = \gamma_{\pm n} \cdot P \) on the power coupled into the cavity. We interpret this result as Raman scattering in the medium giving rise to background emission centered near the pump wavelength. The count rates are several orders of magnitude above the detector dark count rates and they limit the CAR to \( \frac{2}{\pi \tau} \left( \frac{\omega_{0}}{\gamma_{\pm n}} \right) \). We have extracted the formula by assuming a Cauchy distribution of the correlated photon pairs of width \( \tau \) and a binning time much shorter than the correlation time, which is well fulfilled in our experiment (see figure 3(a)). This shows that better background subtraction by spectral filtering at higher orders \( n \) facilitates detection of correlations with higher CAR. For \( n = 2 \) we compute a CAR of \( 3.3 \pm 0.3 \), which is in excellent agreement with the measured values depicted in 3(c).

We also have observed correlated photon pairs in the third order at frequencies \( \omega_{3-} = 2\pi \times 374.831 \) THz and \( \omega_{3+} = 2\pi \times 389.988 \) THz for \( \omega_0 = 2\pi \times 382.410 \) THz and 0.67 W intracavity power. This signals the versatility of our approach to generate photon pairs at controllable frequency difference. In this context, the liquid filling of our cavity offers the unique advantage that continuous changes of the cavity length and hence continuous adjustments of the frequency of the emitted photon pairs are possible. The measured photon rate coefficient in third order is \((0.58 \pm 0.22) \text{ W}^{-2} \text{ s}^{-1}\). In principle, the rate of photon pairs should be independent of the order of the free spectral range. However, in our realization, we have observed some optical damage from the high-intensity experiments in the second order in addition to the lower reflectivity for increasing order of \( n \), with both effects reducing the finesse.

In conclusion, we have demonstrated a liquid-filled microcavity at very high finesse and show that liquids, in addition to their high nonlinearities, can exhibit very low absorption coefficients (comparable to pure SiO₂), which makes them attractive for nonlinear optics studies. We have studied the generation of correlated photon pairs from such a microcavity using SFWM. Photons are emitted in pairs of longitudinal modes equally split from the pump laser frequency and we have detected correlated pairs up to order \( n = 3 \) thereby demonstrating a photon pair source with adjustable frequency spacing between the photons of a pair.
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