Ultrafast redistribution of vibrational energy in liquids
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7 Correlation properties of parametrically generated light

The temporal coherence or correlation time $\tau_c$ of parametrically generated mid-infrared light is determined by measuring the twin-correlation peak in the sum-frequency spectrum as a function of delay between the signal and idler. The correlation time $\tau_c$ of the generated signal and idler fields was found to lie in the picosecond range and follows the frequency-dependence, predicted from the bandwidth by the relation $\tau_c = 1/\Delta\omega$.

7.1 Introduction

In parametric generation, a strong pump field is converted to signal and idler fields. In this process signal and idler start at the zero-photon level, which has as a consequence that the classical description using Maxwell's equations fails. If there are no photons present in a mode of the electric field, the expectation value of the field operator is zero. The expectation value of the square of the electric field operator, on the other hand, is non-zero due to the zero-point energy. The spread in the electric field given by

$$\Delta E = \sqrt{\langle E^2 \rangle - \langle E \rangle^2}$$

is thus also non-zero.44 This means that there is quantum noise present at the signal and idler input fields. These zero-point or vacuum-fluctuations can act as a seed for the parametric generation and subsequent parametric amplification.38 This means that parametric generation, like the Casimir effect,47-48,157 is a macroscopic manifestation of microscopic quantum fluctuations.

The individual phases of the signal and idler fields in parametric generation can obtain any value, because these fields start from quantum noise. However, after being significantly amplified, the sum of their phases is related to the phase of the pump field, because parametric amplification is a phase-sensitive process. This means that the phases of the amplified signal and idler fields can be strongly modulated, leading to a bandwidth given by Equation (2.5), but these modulations will be complementary. As a result of the strong phase modulation, the (spectral) bandwidth of the parametrically generated signal and idler fields can be much larger than than the bandwidth of the pump field.

When the signal and idler fields are recombined in a sum-frequency generation process, generally a broad sum-frequency spectrum will result from the large-bandwidth signal and idler fields. However, when the signal and idler fields overlap within the coherence time of their phase-modulations, these modulations will cancel, resulting in a narrow peak in the sum-frequency spectrum. This narrow peak will have the same width as the pump spectrum. This narrow peak has been observed and is denoted as the twin-correlation peak.1
The term twin-correlation peak has been derived from a quantum-mechanical picture of the parametric generation process. In parametric generation, a pair of photons \textit{(twins)} is generated simultaneously from one pump photon. The occurrence of a narrow peak in the sum-frequency spectrum is thought to signify the recombination of a photon with its twin brother. This picture is not entirely correct as each photon need not be recombined with its twin for the sum-frequency spectrum to display a narrow feature. Photons generated shortly after each other can still give rise to a twin-correlation peak, as the phases of signal and idler fields cannot change infinitely fast, since this would imply an infinitely high frequency.

Measuring the \textit{(relative) intensity} of the twin-correlation peak as a function of delay between the signal and idler fields in the sum-frequency generation when they are recombined, will reveal the correlation time of the parametrically generated fields, which is a measure for the correlation time of the zero-point electric field, which acts as a seed for the parametric generation process. Generally, the coherence time of the phases of the signal and idler fields will be determined by the spectral bandwidth \( \Delta \omega \) of the parametrically generated light. The coherence time or correlation time \( \tau_c \) is related to the bandwidth by \( \tau_c = \frac{1}{\Delta \omega} \), according to the Wiener-Khintchine theorem.\(^{68,121,125}\) This relation has been confirmed in an experiment, measuring the coincidence counts of signal and idler photons.\(^{121,125}\)

The correlation properties of signal and idler fields have been used in a wide range of quantum optical experiments. A pair of twin-photons can be regarded as an \textit{entangled state} and has been used to perform so-called Einstein-Podolsky-Rosen (EPR) experiments. This entangled or EPR-state was proposed to point out the apparent incompleteness of quantum mechanics.\(^{70}\) These \textit{gedanken} experiments were later extended by Bell to demonstrate the incompatibility of quantum mechanics with local descriptions by means of the Bell inequalities.\(^{24,25}\) These inequalities are violated if the local realistic description fails, as was shown in different experiments, using parametric generation.\(^{134,168,169}\) These entangled states, formed by parametrically generated photons, have also been proposed for and used in e.g. "quantum teleportation"\(^{17,18}\) and "quantum cryptography". For a comprehensive treatment of the subject of quantum optics see References 14, 149, 153, 187, and 204.

The twin-correlation peak can also be used as an indicator for or a measure of correlation between parametrically generated fields. If signal and idler photons emanating from the same location in the generating medium are used, only information on the temporal correlation of the zero-point electric field will be obtained. If on the other hand signal and idler photons from two different regions are recombined in the sum-frequency generation, the relative intensity of the twin-correlation peak can be used as a measure for the spatio-temporal correlation of the zero-point electric field.

### 7.2 Experimental Remarks

In these experiments we use experimental set-ups that are based on a set-up for the generation of picosecond mid-infrared pulses which is described in more detail in Section 5.2. These pulses have previously been used in time-resolved non-linear spectroscopy, see e.g. Chapter 5 and References 18, 34, 39, 42, and 210.

In all experiments, pulses from a Nd:YAG laser (Quantel YG502C) are used as pump
pulses in the parametric generation and amplification processes in lithium niobate (LiNbO₃) crystals. This pump laser delivers pulses with a duration of 34 ps (FWHM) and an energy up to 60 mJ per pulse at a wavelength of 1064.1 nm and a repetition rate of 10 Hz. An autocorrelation trace of these pulses, generated through sum-frequency generation in a 6.5 mm BBO crystal, is shown in Figure 7.1. Typical spectra of signal and idler pulses generated in this set-up are shown in Figure 7.2.

7.3 TEMPORAL CORRELATION

7.3.1 EXPERIMENT

The experimental set-up which is shown schematically in Figure 7.3 was used to measure the correlation time of the parametrically generated light as a function of the frequency of the signal and idler pulses. In a LiNbO₃ crystal (5 cm long, optical axis cut at 47.1°) signal and idler pulses are generated by 4 mJ pump pulses. The part of parametrically generated light which travels collinearly with the pump beam is amplified in a second LiNbO₃ crystal. After the second crystal, pump and idler wavelengths are filtered out, yielding signal pulses
with a typical energy of 18 $\mu$J per pulse. These signal pulses are split in two equal parts and used as a seed for two separate amplification stages. The amplified seed pulses are then used in two further amplification stages, each yielding signal and idler pulses with typical energies of 500 $\mu$J and 250 $\mu$J per pulse, respectively. The signal pulses of one amplification stage and the idler pulses of the other are then collinearly combined in a $\beta$-barium borate ($\beta$-BaB$_2$O$_4$ or BBO) crystal. The light generated by the sum-frequency generation process in the BBO crystal is then analysed using a spectrometer with an Optical Multichannel Analyser (OMA). The signal and idler pulses can be delayed with respect to each other using a variable delay in the path of the signal pulse.

The frequency of the signal and idler pulses was varied by tuning the angles of the LiNbO$_3$ crystals. Sum-frequency spectra were recorded at different signal and idler frequencies and at different delay values between the signal and idler pulses.

### 7.3.2 Results and Discussion

Several sum-frequency spectra recorded at different delay values between the signal beam of one amplification stage and the idler beam of the other are shown in Figure 7.4. These spectra were measured with the OPG/OPA stages tuned to a signal frequency $\omega_{\text{signal}}=5830$ cm$^{-1}$ ($\lambda_{\text{signal}}=1715$ nm). When the delay between signal and idler is larger than 2 ps, a broad sum-frequency spectrum, centred around 9398 cm$^{-1}$ (1064.1 nm) is observed. At smaller delay values, a narrow peak appears on top of the broad spectrum. This narrow peak, which is the twin-correlation peak, is at its maximum at delay zero. It should be noted that the twin-correlation peak disappeared when the pinhole (diameter 200 $\mu$m) in front of the beam splitter, which splits the seed in two, was removed.

In Figure 7.5 the intensity of the maximum of the sum-frequency spectra at
Figure 7.4. Sum-frequency spectra at different delays between signal and idler at $\omega_{\text{signal}}=830$ cm$^{-1}$.
\( \omega_{\text{signal}} = 5830 \text{ cm}^{-1} \) at different delay values is plotted as a function of delay between the signal and idler pulses. In order to obtain a good fit of the measured data of these "peak heights" as a function of delay, a Savitzky-Golay filtering procedure\(^{174} \) was applied, as the data was quite noisy. This filtering algorithm was originally developed to extract peak widths and heights from noisy spectrometric data.\(^{182} \) The characteristic feature of this filtering scheme is that the peak widths and heights of the original data are preserved. After the filtering procedure a good fit of a sum of two Gaussians (with a full width at half-maximum of 1.3 ps and 29 ps respectively at \( \omega_{\text{signal}} = 5830 \text{ cm}^{-1} \)) to the data was obtained. In Figure 7.5 this fit is represented by the solid line. The long timescale is identified with the cross-correlation time of the intensity profile of the signal and idler pulses and the short timescale with the correlation time \( \tau_c \) of the electric field.

The results of the experiments at different signal and idler frequencies are shown in Figure 7.6, where the correlation time of the parametrically generated light is plotted as a function of signal frequency. The solid line is calculated using Equation (2.5) for the bandwidth of the generated signal and idler pulses, the Sellmeier dispersion equations for LiNbO\(_3\), see References 41 and 192, and assuming a gain factor \( g_0 \) of 0.4 cm\(^{-1} \) over the length of the last amplification crystal.

Although these measurements do not yield accurate measurements of the correlation time \( \tau_c \) of the parametrically generated light, the expected increase of \( \tau_c \) when tuning away from degeneracy is clearly visible, when comparing the data to the calculated curve in Figure 7.6.

The measured values for \( \tau_c \) also agree with the value of \( \tau_c = 1.13 \pm 0.07 \) ps at \( \omega_{\text{idler}} = 3450 \text{ cm}^{-1} \) (\( \omega_{\text{signal}} = 5950 \text{ cm}^{-1} \)), obtained from previous (incoherent) photon-echo experiments with a slightly modified set-up.\(^{207,210} \) These incoherent photon-echo experiments make use of the fact that the time resolution is determined by the coherence time of the pulses, rather than the pulse duration.\(^{215} \) This has lead to photon-echo experiments
with (sub)-picosecond time resolution with nanosecond laser pulses, but also using a synchrotron and even a light bulb as light sources.

7.4 **Spatio-temporal correlation**

7.4.1 **Experiment**

The experimental set-up shown in Figure 7.3 was modified in order to measure the spatio-temporal correlation of parametrically generated light. This set-up is shown schematically in Figure 7.7. Again, signal and idler pulses are generated in a LiNbO$_3$ crystal. After pump and idler are filtered out, the signal pulses are split into two equal parts and amplified in two separate amplification stages. In front of the beam splitter that divides the signal pulses in two, a lens is placed in such a way that the front face of the crystal is imaged in the planes of pinholes B1 and B2. Pinhole A is intended for alignment purposes. All pinholes had a diameter of 200 μm. Sum-frequency spectra were recorded at different delay values between the signal of one amplification stage and the idler of the other stage.

7.4.2 **Results and discussion**

When only pinhole A is in place in the set-up described Figure 7.7, a twin-correlation peak is observed in the sum-frequency spectra, similar to the results described in §7.3.2. This is not surprising, as the set-up, without the lens or pinholes B1 and B2 present, is in principle the same as the one in Figure 7.3. When pinhole A is subsequently removed, the twin-correlation peak disappears.

When pinholes B1 and B2 are placed with pinhole A present, the twin-correlation peak is present in the sum-frequency spectra, even when pinhole A is removed. When the position of pinhole B1 is varied in a lateral direction, the intensity of the twin-correlation peak
Figure 7.7. Experimental set-up for measuring the spatio-temporal correlation of the parametrically generated light. (Legend: OPG: Optical Parametric Generation, OPA: Optical Parametric Amplification, BS: beam splitter, SWP: Short-wave-pass filter and mirror for 1064 nm, LWP: Long-wave-pass filter and mirror for 1064 nm, SFG: Sum-frequency generation, OMA: Optical multichannel analyser, R1064: mirror for 1064 nm.)

Figure 7.8. Intensity of the twin-correlation peak at different positions of pinhole B1 (see Figure 7.7, without the presence of a lens.)
Table 7.1. Presence of twin-correlation peak with different combinations of optical elements present in the set-up shown in Figure 7.7.

<table>
<thead>
<tr>
<th>Elements in place</th>
<th>Twin-correlation peak present?</th>
</tr>
</thead>
<tbody>
<tr>
<td>none</td>
<td>no</td>
</tr>
<tr>
<td>pinhole A</td>
<td>yes</td>
</tr>
<tr>
<td>pinholes B1,B2</td>
<td>yes</td>
</tr>
<tr>
<td>pinholes A,B1,B2</td>
<td>yes</td>
</tr>
<tr>
<td>lens</td>
<td>no</td>
</tr>
<tr>
<td>lens and pinholes B1,B2</td>
<td>no</td>
</tr>
<tr>
<td>lens and pinhole A</td>
<td>yes</td>
</tr>
</tbody>
</table>

With respect to the background at delay zero changes, as can be seen in Figure 7.8. However, these data do not provide information on the spatial correlation of the parametrically generated light or the zero-photon field in the generating crystal. As there is no imaging without the lens in place, the light going through pinhole B1 (and B2, for that matter) cannot be related to a certain position in the front face of the generating crystal. The measured pattern, is therefore only providing information on the correlation properties or “speckle” across the beam profile of the generated signal beam.

When the lens was placed with pinholes B1 and B2 in place, no correlation peak was observed. With the lens in place, the twin-correlation peak was only observed when pinhole A was in place as well. A summary of the presence or absence of the twin-correlation peak in combination with the different elements can be found in Table 7.1.

In order to get a good spatial resolution for the measurement of the spatio-temporal correlation, one needs to gather parametrically generated light with as many directions as possible. The signal and idler fields that are generated non-collinearly with the pump beam and that propagate in an off-axis direction contribute to the spatial resolution. However, these fields have a non-zero angle between signal and idler and will be phase-matched at different frequencies than the collinearly propagating signal and idler fields. Therefore, the off-axis generated fields hold different “information” on frequency and phase than the collinearly generated beams. In the sum-frequency generation, only signal and idler with complementary frequencies will contribute to the twin-correlation peak, because then the phase modulations cancel. The fields that were generated with a different frequency will contribute to a broad background in the sum-frequency spectrum.

If only the collinearly generated signal and idler fields were amplified and subsequently used in the sum-frequency generation, only the twin-correlation peak would be observed, without a broad background. The more of the other off-axis fields are “mixed in”, the more intense the broad sum-frequency spectrum will become. The necessity of the presence of the pinhole before the beam splitter in in set-ups in both Figure 7.3 and Figure 7.7 can be explained in this way. Without the pinhole to spatially select the parametrically generated light travelling collinearly with the pump, the eventual broad background will become so intense that the twin-correlation peak is completely “drowned”. Indeed, the intensity of the mid-infrared pulses increases with the pinhole removed, as does the total intensity of the sum-frequency light.
An additional indication of the phase-sensitivity of the processes giving rise to the occurrence of the twin-correlation peak is provided by some observations concerning the sum-frequency generation. When the signal and idler beams are focused by a lens and/or overlapped in a non-collinear geometry, no twin-correlation peak is observed. Only when the signal and idler beam are combined collinearly without focusing in the BBO crystal, the twin-correlation peak is present around delay zero. It is very likely that in any other geometry than the collinear one, the parts of either beam that give rise to the twin-correlation peak are either smeared out or simply do not overlap in the crystal. This again will lead to a disappearance of the twin-correlation peak, because it is either drowned or absent.

The need for spatial resolution for the observation of the twin-correlation peak is in strong contradiction with the presence of an angular frequency dependence. The twin-correlation peak can therefore not be used as a measure for spatial correlation in these experiments. The solution to circumvent these problems could be to use very thin crystals in the parametric generation process. However, in this case the intensity of the generated fields will be very low.

### 7.5 Directional correlation

The experiment described in this section was devised in order to test whether parametrically generated light with different directions, but with no phase-mismatch can give rise to a twin-correlation peak. If two pump beams are overlapped non-collinearly and are aligned in such a way that they make equal angles with the normal to the crystal face, the two collinearly generated signal fields are expected to have the same frequencies and phase-(mis)match and a twin-correlation peak is expected to be observed. In this way, the type of correlation that is sampled is not the spatial correlation but the directional correlation of the parametrically generated light.

#### 7.5.1 Experiment

The set-up that was used in order to sample directional correlation of the parametrically generated light is shown in Figure 7.9. Here, two pump beams are overlapped non-collinearly in a LiNbO₃ crystal. These pump beams are aligned in such a way that they overlap on the front face of the crystal and that they make equal angles with the normal to the crystal face. The signal beams from the parametrically generated light in the first crystal are subsequently amplified. Again, sum-frequency spectra were recorded at different delay values between the signal of one amplification stage and idler pulses of the other stage.

#### 7.5.2 Results and discussion

By overlapping the two pump beams in the front face of the generating crystal in the experimental set-up described in Figure 7.9, the same volume of the crystal can be “probed” by these two pump beams, without the need for imaging optics. This would in theory circumvent the problems encountered in the previously discussed imaging experiments. If the parametrically generated light, generated by the two pump beams were seeded by the same quantum fluctuation, a twin-correlation peak could be expected, and by varying the overlap of the two pump beams, measurement of the spatial correlation would be possible.
No twin-correlation peak was observed in this configuration, which suggests that there is no directional correlation of the quantum fluctuations in the LiNbO$_3$ crystal.

7.6 Conclusions

The temporal coherence of parametrically generated mid-infrared light was determined by measuring the twin-correlation peak in the sum-frequency spectrum as a function of delay between the signal and idler. Due to the phase-sensitive nature of the parametric generation and amplification processes, the determined coherence or correlation time $\tau_c$ can be directly related to the $\tau_c$ of the zero-point electromagnetic field, from which parametric generation starts. The correlation time $\tau_c$ of the generated signal and idler fields was found to lie in the picosecond range and was found to follow the frequency-dependence, predicted by the bandwidth from the relation $\tau_c = 1/\Delta \omega$.

The proposed method for the determination of the spatio-temporal correlation by using the twin-correlation peak as a measure of coherence was found to be unsuited. The need for a high spatial resolution and therefore a wide acceptance angle of the parametrically generated light was found to oppose the observation of a twin-correlation peak, as the frequency of the parametrically generated light shows a strong angular dependence.