Generation of intense sub-picosecond pulses in the mid-infrared

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We describe the use of difference-frequency generation, by mixing two intense femtosecond pulses in a 5-mm long BBO crystal, to generate high power 2.5 μm pulses with a duration of less than 0.5 ps. The pulses are generated by mixing the output of an amplified CPM laser with an amplified part of a continuum. The generated pulses have an energy of 4 μJ which amounts to a photon-conversion efficiency of 5 percent.

1. Introduction

The generation of ultrashort infrared pulses is among others of interest to many workers studying time-resolved vibrational dynamics of molecules and to scientists involved in the time-resolved study of semiconductors. In the past, several techniques such as parametric generation and amplification [1-4], parametric oscillation [5] and difference-frequency generation [6-8] have been used to generate tunable picosecond infrared pulses.

However, many dynamical processes take place on a sub-picosecond timescale and there clearly exists a need for infrared pulses of sub-picosecond duration. During the last decade, in order to generate tunable femtosecond infrared pulses, one has used difference-frequency generation, by mixing the output of an amplified CPM laser and part of a generated white-light continuum [9-11], parametric oscillation [12] and parametric amplification [13]. Recently Laenen et al. [14,15] reported on the generation of femtosecond infrared pulses by a synchronously pumped parametric oscillator. In another report [16], light of a travelling wave dye-laser was mixed with part of a white-light continuum to generate femtosecond pulses between 1 and 10 μm. All these techniques are well suited for purposes where tunability is needed, but they all produce relatively low power pulses. A somewhat different approach was recently demonstrated, in which femtosecond pulses with a fixed wavelength of 1.55 μm were amplified to the microjoule level using a NaCl F-center crystal [17]. In this report we show that it is possible to generate high-power infrared pulses by mixing the output of an amplified CPM laser with the amplified part of a generated femtosecond continuum in a 5 mm long beta barium borate crystal (BBO) to generate their difference frequency.

2. Experimental

The experimental setup used to generate the infrared pulses was built around a CPM-laser that has a repetition rate of 110 MHz and is pumped by a Spectra-Physics 2016 argon-ion laser. The CPM is stabilized using a feedback loop that couples the output of the CPM to the pump power of the argon laser [18]. The CPM operates at 620 nm and has an average cw-power of 10 mW. The pulses are amplified at a 10-Hz repetition rate in four Bethune-type [19] dye cells that are filled with a solution of Sulfo-rhodamine 640 in methanol. The amplifier, pumped by synchronized nanosecond pulses from a frequency-doubled, injection-seeded Spectra Physics GCR-4 Nd:YAG laser, raises the energy per pulse to a 100 μJ. The next stage of the setup is depicted schematically in fig. 1. About 40 μJ of the amplified pulse is used for continuum generation in a 2.5 cm long cell, filled with water. An interference filter with a band-
width of 10 nm centered around 825 nm was used to select a portion of the continuum for further amplification in three Bethune-type dye-cells filled with LDS 821 dye (785–850 nm) dissolved in methanol. After amplification, the 825-nm pulses had an energy of approximately 40 μJ. The pulse duration as measured by standard autocorrelation by second-harmonic generation in a 5-mm KDP crystal was found to be 220 fs, based on the assumption that the pulses have a shape corresponding with a sech² (fig. 2). We assume that the bandwidth of the pulses remains approximately 10 nm, determined by the width of the interference filter, in view of the assumption that the gain is not very wavelength dependent within this interval in the centre of the gain curve of the dye. The remaining 60 μJ of the 620-nm pulses was amplified once more to an energy of 300 μJ per pulse. The 620-nm pulses had a bandwidth (fwhm) of ≈4.5 nm implying that the pulses were not far from the bandwidth limit for a sech². The 620-nm pulses were sent through a Fresnel-rhomb to rotate their polarization by 90 degrees. The pulse duration as measured by autocorrelation was found to be 200 fs, with
an autocorrelation signal similar to that of the 825-nm pulse.

After passing through a variable delay, the amplified 620-nm pulse and the 825-nm pulse were combined collinearly using a dichroic beamsplitter. Both beams had a diameter of 2 mm. Overlap in time was found by crosscorrelating the two pulses, using sum-frequency generation in a short KDP crystal. In order to generate the infrared pulses, the KDP crystal was subsequently replaced by a 5-mm long BBO crystal for type I difference-frequency generation at a phase-matching angle of 20.3 degrees, as calculated using the Sellmeier’s equations for BBO [20] \((620 \text{ nm}(e) - 825 \text{ nm}(o) + 2.5 \mu m(o))\). Directly behind the crystal, the generated 2.5-μm pulse is transmitted by a silicon dichroic beamsplitter that reflects the 620-nm and 825-nm pulses. The infrared power was maximized by measuring the infrared signal with a PbSe photo-conductive cell as a function of the delay between the 620-nm and the 825-nm pulses. The average power of the infrared pulses was measured with a Molectron 53-05 pyroelectric power meter. A measure for the pulse duration of the infrared was obtained by type II sum-frequency generation of the pulse with the remainder of the 620-nm pulse in a 5-mm long BBO crystal, at a calculated phase-matching angle of 23.9 degrees. \((2.5 \mu m(e) + 620 \text{ nm}(0) \rightarrow 497 \text{ nm}(e))\). To guide the infrared beam, aluminium- and gold-coated mirrors were used. The infrared absorption of the BBO crystal was determined with a conventional Perkin-Elmer infrared double-beam spectrometer.

### 3. Results and discussion

In order to obtain the pulse duration of the infrared pulse we cross-correlate the 2.5-μm pulse with the remainder of the 620-nm pulse. The sum-frequency signal is shown in fig. 2 as a function of delay between the 2.5-μm and the 620-nm pulses. The curve is a fit based on a sech² pulse shape that corresponds with a sech² but should be considered a guide to the eye. We did not measure the bandwidth of the infrared pulse.

We arrive at an estimate of the infrared pulse duration of less than 0.5 ps as will be explained next.

Generally, in nonlinear processes like difference-frequency and sum-frequency generation, the group velocities of the interaction pulses are not equal. This means that when the two pulses propagate through a long crystal, temporal overlap will worsen or improve depending on the initial delay between the pulses. It implies that a long cross-correlation signal may have two possible causes. It can be caused by either the infrared generation process or by the measurement of the cross-correlation signal itself (or both). The former can be explained by noting that if the generating pulses in difference-frequency generation have approximately equal group velocities but different from the group velocity of the generated signal, the latter may lag behind or advance with respect to both generating pulses. Because new light is continuously being generated, the generated pulse will become much longer than the original pulses, the final pulse duration depending on the crystal length.

The measurement itself can also be responsible for a long crosscorrelation signal. This can be seen by noting that if the two fundamental pulses in sum-frequency generation have different velocities, the curve that represents the generated signal as a function of initial delay between the two pulses is broadened. This makes a precise determination of the pulse duration difficult.

In the case of BBO we calculated the group velocities for the interacting pulses from

\[
v_g = \frac{c}{n + \omega \frac{dn}{d\omega}}.
\]

The calculated results for the type I difference-frequency generation process and the type II sum-frequency cross-correlation process are listed in table 1. For the type II process there are significant differ-

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>Type I travel time</th>
<th>Type II travel time</th>
</tr>
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<tbody>
<tr>
<td>497 nm</td>
<td>-</td>
<td>84 fs</td>
</tr>
<tr>
<td>620 nm</td>
<td>0 fs</td>
<td>0 fs</td>
</tr>
<tr>
<td>825 nm</td>
<td>-46 fs</td>
<td>-</td>
</tr>
<tr>
<td>2.5 μm</td>
<td>-42 fs</td>
<td>-643 fs</td>
</tr>
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| Table 1 Calculated times needed for the interacting pulses to travel through 5 mm of BBO, with respect to the 620-nm pulse. A negative number means that the pulse travels faster than the 620-nm pulse. Type I: 620 nm (e) → 825 nm(o) + 2.5 μm(o). Type II: 620 nm(o) + 2.5 μm(e) → 497 nm(e). |
ence in travel times between the 620-nm and 2.5-μm pulses. These numbers seem to indicate that the origin of the fact that the crosscorrelation signal is broader than the 800-nm and 620-nm autocorrelation signals lies in the measurement of the pulse duration itself. However, we note that the numbers given for the group velocities should be viewed with some suspicion, since the Sellmeier equations from which they were calculated were fitted to refractive index data obtained below 1000 nm [21]. This casts some doubt on their usefulness for longer wavelengths. It is for this reason that the IR-pulse duration of 0.5 ps that we extracted from the crosscorrelation signal, must be considered an upper limit.

The infrared absorption as a function of wavelength as measured with the infrared spectrometer was found to be similar to that in ref. [21]. The transmission of the 5-mm long BBO crystal at a wavelength of 2.5 μm has a value of 55 percent. The infrared absorption will have some negative influence on the efficiency of the difference-frequency generation process. The average energy of the 2.5-μm pulses was found to be 4 μJ. This amounts to a photon-conversion efficiency of 620 nm→825 nm + 2.5 μm, of 5 percent. Taking the upper limit on the pulse duration of τp=0.5 ps we estimate a lower limit on the infrared pulse power of ≈10 MW.

The divergence of the infrared beam was found to be ≈1.5 mrad. The small divergence of the generating 620-nm and 825-nm beams clearly is advantage if one wants to have a low-divergence infrared beam. This is contrary to the case of parametric generation, where using for example only the 620-nm pulse as a pump, the generated infrared emerges as a cone from the exit face of the crystal. In that case a second crystal at a considerable distance from the first one is needed for amplification to reduce the divergence.

The tunability of the setup is only limited, in principle, by the availability of dyes that emit in the appropriate wavelength region and by the infrared absorption in BBO for wavelengths larger than ≈2.5 μm. Tunability can be achieved quite easily by choosing the appropriate wavelength within the emission bandwidth of the LDS dye, with suitable interference filters. In this way intense infrared pulses between 2.3 μm and 2.9 μm can be generated fairly easily.

Finally, we would like to point out that BBO is a crystal to be favoured over many other non-linear crystals with a high second-order susceptibility like LiNbO3, because the dispersion n(ω) in the wavelength region of interest (i.e. 620–2500 nm) is relatively small, allowing the use of longer crystal lengths before group-velocity differences become too large.

4. Conclusions

We demonstrated that it is possible to generate intense mid-infrared sub-picosecond pulses in a 5-mm long BBO crystal by difference-frequency mixing of two amplified femtosecond pulses. The high infrared intensities that can be realized with this setup may find numerous applications in the sub-picosecond time-resolved study of many materials.

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