Chapter 2

Generation and characterization of tunable mid-infrared femtosecond pulses

Abstract

In this Chapter we describe the design of an optical parametric amplifier that allows the generation of infrared laser pulses suitable for ultrafast nonlinear spectroscopy on the OH-stretching mode of water molecules. Mid-infrared femtosecond pulses, tunable in the range of 2800-3800 cm\(^{-1}\) with sub-100 fs duration and ~15 µJ energy are produced by an optical parametric amplifier driven at 1 kHz by 750 mJ, 800-nm pulses from a Ti:Sapphire amplifier. In this setup, tunable low-energy femtosecond pulses in the near infrared are generated by continuum amplification in \(\beta\)-barium borate, and subsequently the near-infrared pulses are amplified in two stages in potassium titanyl phosphate pumped by 800 nm radiation to produce intense mid-infrared pulses. A novel method is used to characterize the ultrashort IR pulses. The technique utilizes a frequency-resolved pump–probe geometry common in the applications of ultrafast spectroscopy.

Part of the work as presented in this Chapter is covered by the following papers:


2.1 Introduction

Nonlinear infrared (IR) spectroscopy of the condensed phase is, at present, one of the most dynamic and developing fields in physical chemistry. The first experiments in this area employing the output of free electron lasers have revealed subpicosecond timescales of vibrational dynamics in liquid phase [1-3]. In the last few years, femtosecond mid-IR pulses have been rapidly gaining importance for site-sensitive vibrational spectroscopy on condensed matter [4-9]. Thus, generation and characterization of tunable femtosecond laser pulses in the mid-IR region is currently a subject of great importance.

There are no direct femtosecond laser sources for the generation of tunable pulses with a spectrum centered at wavelengths longer than 3 µm due to the absence of suitable laser materials and mode-locking techniques. Therefore, techniques based on the processes of parametric frequency down-conversion are usually employed. Such methods often offer high conversion efficiency and very broad tunability of the output radiation. The progress in engineering of relatively simple and versatile frequency conversion systems for the generation of high-energy femtosecond pulses, tunable throughout the visible and IR regions, was greatly stimulated by advances in the development of Ti:Sapphire lasers and amplifiers [10-15].

In the visible region generation of tunable femtosecond pulses with an energy of several micro-Joules has been achieved by making use of BBO based parametric oscillators and amplifiers. Employing non-collinear schemes with a BBO based parametric amplifier seeded with white light, allowed generation of pulses with spectra spanning from the near ultraviolet to the near-IR region and having sub-5 fs duration [16,17].

In the mid-IR region, subpicosecond pulses suitable for nonlinear spectroscopic experiments have been generated employing a number of different nonlinear crystals and several schemes. For the generation of the IR pulses in the spectral range of 2-4 µm a KTP (KTiOPO₄) crystal is commonly used. However, it was recently shown that utilizing aperiodically poled lithium niobate (LiNbO₃) allows generation of mid-IR pulses in the 3 µm region with duration as short as 50 fs [18]. Also a noncollinear geometry for parametric interaction in MgO:LiNbO₃ has been employed to produce high energy pulses in the 3 µm region with a duration of about 125 fs [19]. For down conversion further to the mid-IR (down to 10 µm) nonlinear crystals such as AgGaS₂ or HgGa₂S₄ are usually used [20-24].

The methods of frequency down-conversion are traditionally divided into those employing the principles of the optical parametric oscillator (OPO) and the optical parametric amplifier (OPA). In the first case, the nonlinear crystal is placed in a resonator cavity that provides high efficiency of frequency conversion. In the parametric amplification technique, the frequency conversion occurs during one pass through the nonlinear medium. The latter method has sufficient efficiency for ultrashort pulses with high peak intensities. In order to increase the efficiency of the parametric process the parametric amplifier is usually seeded with a relatively weak pulse, which is amplified. Thus, the techniques of parametric amplification can be classified by the methods of seeding. An output of an optical parametric oscillator or amplifier can be used to seed another parametric amplifier, forming in such a way nonlinear frequency conversion process in several stages. For example, in a narrow region around 3-µm, nearly transform-limited 0.5 µJ pulses at a repetition rate of 205 kHz with duration of about 160 fs were generated by parametric amplification in noncritically phase matched KTP, seeded by a synchronously pumped OPO [25]. A combination of near-IR (NIR) OPO based on KTP crystal and mid-IR OPA with several AgGaS₂ crystals driven by pulsed Nd:YLF laser system allowed generation of 0.5 ps pulses in the range of 2.6-7 µm [26]. Another example of such an approach is that a commercial OPA system (TOPAS) is used to
generate a NIR beam for seeding of a KTP producing mid-IR pulses in the 3 \( \mu \text{m} \) region with a duration of 150-250 fs \([27]\). Quasi-cw laser radiation can also be used for seeding. For example, sub-100 fs pulses tunable in the region of 2.5 - 4.2 \( \mu \text{m} \) were produced in a single stage parametric amplification in the phase-matched KTP crystal pumped by a tunable high-power femtosecond Ti:Sapphire laser at 1-kHz repetition rate and seeded by tunable quasi-cw radiation from a Nd:YAG laser \([28,29]\). In the most recent schemes a white light continuum generated by the self-phase modulation process is often used as the seed \([24,30]\). A relatively simple but efficient method was demonstrated in which amplified, in a double stage BBO OPA near-IR continuum was used to seed a KTP based parametric amplifier generating 200 fs pulses with an energy reaching 13 \( \mu \text{J} \) in the region of 2.5-4.2 \( \mu \text{m} \) \([30]\).

The 3 \( \mu \text{m} \) region is one of the most interesting for research, since such spectroscopically important vibrations as fundamental stretching modes of OH-, NH-, and CH-groups, are situated here. In the current work we focus our attention on the OH-stretch vibration of water molecules in the liquid phase. The spectrum of this vibrational mode is centered at 3400 cm\(^{-1}\). Its full width at half maximum is about 250 cm\(^{-1}\). The fastest dynamics theoretically predicted for this vibration in liquid water has a sub-100 fs timescale \([31,32]\). Sub-100 fs pulses with a spectrum tunable in 3-\( \mu \text{m} \) region are, therefore, required for carrying out nonlinear spectroscopic experiments on this object.

We present here a design of an optical parametric amplifier that allows generation of mid-IR pulses tunable in the range of 2.5-3.5 \( \mu \text{m} \) with energies up to 15 \( \mu \text{J} \) and duration of about 70 fs. The IR pulses generated in the OPA are characterized using a novel technique for pulse characterization. The method is based on the measurement of the frequency resolved pump-probe in a transparent medium with instantaneous nonlinearity. We propose an original algorithm that allows for the efficient pulse retrieval. The algorithm adjusts itself to the pulse complexity thus reducing substantially the processing time.

### 2.2 Generation of the mid-IR femtosecond pulses

For the generation of IR pulses with the required characteristics, we chose a design based on parametric amplification in a KTP crystal. The mid-IR pulses are generated as a difference frequency between a near-IR seed having a wavelength in the range of 1.08-1.18 \( \mu \text{m} \), and the 800 nm pump. The pumping source is a femtosecond Ti:Sapphire laser-amplifier system. The seed pulses are generated in a collinear BBO-based OPA seeded with white light. The tunability of the mid-IR output is achieved by tuning the wavelength of the near-IR seed pulses, the mutual delay between the seed and pump pulses, and the orientation of the nonlinear crystal.

KTP has several advantages over other nonlinear crystals suitable for parametric generation in the 3-\( \mu \text{m} \) region. It has a relatively large damage threshold, a property, that is important to produce the high-energy output necessary for carrying out nonlinear spectroscopic experiments \([28,33]\). Moreover, in the wavelength region around 3-\( \mu \text{m} \), a low group velocity mismatch between the signal-pump and the idler-pump waves can be achieved in this type of crystal \([29]\). This, together with its relatively high nonlinearity, provides high efficiencies of frequency conversion and consequently allows supporting substantial spectral bandwidth in the generated IR pulses.
The KTP crystal is biaxial and the highest effective nonlinearity is achieved in the case of Type-II interaction in the x-z plane \([29,33]\). The phase-matching angle curve for Type-II interaction \([o(pump)-e(signal)+o(idler)]\) is presented in Fig.2.1 \([34]\). The shaded contour in Fig.2.1 depicts the range of crystal orientations employed in our work. The dotted line shows the cut angle of the crystal.

Fig.2.1. The phase-matching angle curve for Type-II interaction \([o(pump)-e(signal)+o(idler)]\) in KTP with a pump wavelength of 800 nm. Solid line corresponds to the signal wave, while the dashed line to the idler. The shaded contour depicts the range of phase-matching angles used in this work. The dotted line shows the cut angle of the crystal.

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The schematic of the OPA system is shown in Fig.2.2. The characteristics of the parametric amplifier are summarized in a table shown in Fig.2.2. The setup is pumped by the output of a multipass Ti:Sapphire amplifier providing \(\sim 30\) fs pulses centered at 800 nm with an energy of \(\sim 750\) µJ. The system is driven at a 1 kHz repetition rate. The pump beam diameter is \(\sim 1\) cm. Thus, its intensity reaches \(0.1\) TW/cm\(^2\). Due to such high intensity the pump beam undergoes substantial self-phase modulation in any optical material leading to alteration of its spectral and phase characteristics \([35]\). Such modification of the pump pulse can substantially lower the efficiency of the parametric generation process and lead to unpredictable characteristics of the IR pulses generated. Therefore, only reflective optics and thin (300 µm) beam-splitters and \(\lambda/2\) plates (CVI) are employed for the pump beam to minimize the self-phase modulation effect, additionally preventing lengthening of the pump pulses due to group velocity dispersion.
The schematic of the OPA system. DL, mechanical delay line; BS, beam-splitter; L, lens; DM, dichroic mirror; FM, focusing mirror; λ/2 plate; RF, rejection filter. The characteristics of the parametric amplifier are summarized in the table.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Nonlinearity</th>
<th>Material</th>
<th>Cut angle (deg)</th>
<th>Pump wavelength (nm)</th>
<th>Pump energy (μJ)</th>
<th>Generated wavelength (μm)</th>
<th>Energy conversion efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\chi^{(3)}$</td>
<td>Sapphire</td>
<td>-</td>
<td>800</td>
<td>0.1</td>
<td>0.64</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>$\chi^{(2)}$</td>
<td>BBO</td>
<td>30</td>
<td>400</td>
<td>40</td>
<td>1.1</td>
<td>~1</td>
</tr>
<tr>
<td>3</td>
<td>$\chi^{(2)}$</td>
<td>KTP</td>
<td>42</td>
<td>800</td>
<td>150</td>
<td>3</td>
<td>20</td>
</tr>
<tr>
<td>4</td>
<td>$\chi^{(2)}$</td>
<td>KTP</td>
<td>42</td>
<td>800</td>
<td>250</td>
<td>3</td>
<td>25</td>
</tr>
</tbody>
</table>

The parametric amplifier consists of four main stages. In the first stage, a single filament white light continuum is generated by focusing a small portion of the 800 nm radiation (~100 nJ) into a 2-mm sapphire plate. In the second stage the continuum is amplified in a 2-mm BBO crystal (Type I phase-matching, $o$(pump)$-e$(seed)$-e$(idler) interaction, cut angle 30°), and pumped collinearly by the second harmonic of the 800 nm beam. About 200 μJ energy of the fundamental 800 nm beam is employed at this stage. The second harmonic for the pump is generated in a 0.5 mm thick BBO crystal (cut angle 29°) with a conversion efficiency of approximately 20% that provides ~40 μJ of 400 nm radiation. The white light
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The continuum is focused to the parametric amplification crystal by the lens, L2. The pump beam diameter in the nonlinear medium is adjusted by means of a concave focusing mirror (FM1) in such a way that only its central part, where the beam has nearly perfect Gaussian spatial intensity distribution, takes part in the amplification process. This provides high spatial quality of the generated beams. Furthermore, such alignment reduces influence of the spatial walk-off effects. The focal point of the 400 nm beam is ~5 cm behind the nonlinear crystal. The BBO crystal orientation and the mutual delay between the pump and seed pulses are tuned in such a way that a part of the continuum in the range 620-660 nm is amplified. The amplified pulses typically have spectral width of ~25 nm (FWHM) and an energy of the order of 200 nJ. Consequently, idler pulses with ~100 nJ energy, ~250 cm⁻¹ bandwidth and a spectrum positioned in the range 1.1-1.2 µm are generated. The near-IR output is split off the pump and seed beams by means of a dichroic mirror DM2 (CVI) and collimated with a telescope constructed of a positive and a negative lens.

In the third stage the near-IR output is amplified in a 2-mm long KTP crystal (Type II phase-matching, \(o(pump) - e(seed) - o(idler)\) interaction, cut angle 42°) pumped collinearly with 150 µJ of the fundamental beam at 800 nm. The pump beam polarization is preliminarily rotated by 90° by means of a 300-µm half-wave plate. The signal beam has a diameter of ~2 mm, while the pump beam diameter in the nonlinear crystal is adjusted by a concave focusing mirror (FM2) in a similar way as in the first amplification stage. Only the central part of the 800 nm beam, where the spatial intensity distribution approaches perfect Gaussian shape (approximately 10% of the total beam diameter), is employed in the frequency conversion process. The signal gain at this stage equals approximately 30. The energy of the produced near IR pulses is about 3 µJ. Taking into account that the interaction occurs only in the beam overlap area the efficiency of conversion of the pump pulse energy into the signal and idler reaches 20%. Due to the high efficiency of the parametric process the signal amplification is in the saturation regime, i.e. the pump pulse energy is substantially depleted. That effectively broadens the spectrum of the amplified pulses and provides better pulse-to-pulse stability in

![Normalized intensity vs. Wavelength](image)

**Fig.2.3.** An example of the signal spectrum before (dashed line) and after the second amplification stage (solid line).
the output [24]. An example of the signal spectrum before and after the second amplification stage is shown in Fig.2.3 as dashed and solid lines respectively. The duration of the signal pulses, determined from cross correlation measurements with fundamental 30 fs 800 nm pulses at the output of this stage, is approximately 150 fs.

The third amplification stage is constructed analogously to the second one. The near-IR signal and 800 nm pump beams are combined on a dichroic mirror DM3 (CVI) and injected collinearly in a second 2 mm long KTP crystal. The phase matching conditions and interaction type in the nonlinear crystal are the same as those described in the previous stage. The pump energy is \( \sim 250 \mu \text{J} \). The depletion of the pump reaches \( 60 \mu \text{J} \), which corresponds to almost 25% efficiency in energy conversion. Thus from the pump beam depletion the energy of the produced mid-IR pulses can be estimated. In this case it is of the order of 15 \( \mu \text{J} \) (for the spectrum centered at 2.8 \( \mu \text{m} \)). Owing to the large spectral width of the seed pulses (see Fig.2.3) the tunability in the region of our interest 3200 – 3600 cm\(^{-1}\) (for spectroscopy on OH-stretch vibration of HDO molecules) is achieved by changing only the orientation of both KTP crystals and adjusting the mutual delay between the pump and signal pulses. Examples of the spectra of generated mid-IR pulses are shown in Fig.2.4.

After the second amplification stage, the mid-IR pulses are collimated with a CaF\(_2\) lens. The IR radiation is filtered out of the pump pulses by means of an uncoated GaAs plate serving as a low pass filter.

### 2.3 Frequency-resolved pump–probe characterization of femtosecond IR pulses

Precise knowledge of the amplitude and phase of the IR-pulses is required to unravel dynamical processes in molecular systems, which often occur on a time scale comparable to the pulse duration.
Frequency-resolved optical gating (FROG) [36,37] is widely used for ultrashort pulse characterization. A number of outstanding features such as experimental simplicity, uniqueness of the retrieved amplitude and phase, and independent data consistency checks, based on the use of the so-called marginals [38], make FROG an invaluable tool in ultrafast spectroscopy. However, there are several limitations that hamper direct application of conventional FROG techniques in the mid-IR region. For instance, second-harmonic generation (SHG) FROG requires a specifically cut nonlinear crystal and detection in a spectrally shifted region [39,40]. The $\chi^{(3)}$-based variations of FROG (such as self-diffraction or transient-grating) call for a high magnitude of nonlinearity, appreciable pulse intensity, and high detector sensitivity, which are not readily available in the IR. Cross-correlation modifications of the FROG technique (XFROG) [41] necessitate addition of a reference pulse at a different frequency with a duration similar to that of the pulse to be characterized.

Optical heterodyne detection provides a number of advantages over conventional homodyne-based FROG schemes [42,43]. The signal enhancement achieved as a result of heterodyning makes it possible to characterize low-intensity pulses using 3rd-order nonlinearity. The latter in many cases provides perfect phase matching conditions such as, for instance, in the geometry of frequency-resolved pump–probe (FRPP), which is widely used in ultrafast spectroscopic experiments [44]. It is well known that FRPP signals that arise from the electronic response in both transparent and absorbing media within the temporal overlap of the pump and probe pulses (conventionally referred to as “coherent artifact”), are highly sensitive to pulse parameters [43,45,46]. It was recently suggested that analysis of FRPP spectrograms might be suitable for the characterization of ultrafast pulses [43]. However, no practical method has been reported so far.

Here we explore FRPP as a method for IR pulse characterization. The schematic of the FRPP setup is depicted in Fig. 2.5. The output of the OPA is split into two unequal parts by means of an uncoated CaF$_2$ plate. Consequently, the probe beam energy is approximately 4% of the pump. The mutual delay between the pump and probe pulses is controlled by means of a high precision motorized delay line (DL). The pump and probe beams are focused into the sample and recollimated with two 10 cm mirrors. The probe is spectrally dispersed through a ¼-m monochromator (CVI) and its spectral components measured with a liquid-nitrogen cooled InSb detector. A synchronous 500 Hz chopper was inserted in the pump beam. The
pump-probe signal (modulation of the probe beam intensity $\Delta I_{\text{probe}}$) is processed with a lock-in amplifier, while the reference signal (the probe beam intensity $I_{\text{probe}}$) is simultaneously detected as the dc component of the detector output. Both components were digitized and stored in a computer. The difference absorption signal is calculated as the ratio $\Delta I_{\text{probe}}/I_{\text{probe}}$.

The contribution, induced by both pulses, can be expressed as follows [44]:

$$S_{\text{FRPP}}(\Omega, \tau) \propto -\text{Re} \left[ E^*(\Omega) \int |E(t+\tau)|^2 E(t)e^{i\alpha t} dt \right]$$

(2.1)

where $\tau$ is the delay between the excitation and probe pulses, $E(t)$ and $|E(\Omega)|^2$ denote the pulse electric field and spectrum, respectively.

Figure 2.6 presents several examples of FRPP traces of different pulses. Note, that unlike a conventional FROG signal, a FRPP trace comprises both positive and negative values. As is usually the case for other $\chi^{(3)}$-based FROG techniques [38], the FRPP traces, in general, exhibit no mirror symmetry with respect to the time-delay axis, which enables unambiguous determination of the time-flow direction. Furthermore, FRPP offers highly intuitive traces: a curve drawn along the crests follows the group delay, i.e. the first derivative of the spectral phase (white curves in Fig.2.6). The sign of the 2nd order phase derivative (group delay dispersion, GDD) determines whether the white curves in Fig.2.6 follow the crests of positive or negative segments of the FRPP pattern. Intervals with a zero GDD value run through zero-magnitude FRPP elements. In particular, the trace corresponding to a spectrum-limited pulse, consists of four distinct quadrants with well-defined boundaries (Fig.2.6a). Because of intrinsic phase-matching, the demand for a thin nonlinear medium, as is the case in SHG or SD FROG, is substantially relaxed in FRPP.
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Fig. 2.7. Experimental (a) and reconstructed (b) FRPP traces and the results of the pulse retrieval in the frequency (c) and time (d) domains. Shaded contours in (c,d) depict the pulse intensities while open circles represent the phase. Independently measured pulse spectrum is shown in (c) by solid dots.

The proposed technique essentially requires a short nonlinear length (defined as $L_{NL} = c/\alpha n_2 I$) in contrast to other methods based on self-phase modulation [47,48]. Consequently, FRPP has several important advantages over these methods, including reduced material dispersion, absence of tight requirements on the transversal intensity distribution, and retains intuitiveness of the traces. Other essential constraints on FRPP concern the 3rd-order nonlinearity: its electronic (instantaneous) part should dominate over two-photon absorption [49] and Raman contributions [37,50]. Both conditions can usually be fulfilled by the proper choice of the nonlinear material. In particular, materials in which Raman modes are inactive (for instance, alkali halides [51]) show a great potential in this respect. An essential and powerful feature of the FROG technique is the ability to check experimental data via the marginals [38]. The peculiarity of FRPP lies in the fact that both temporal and spectral marginals (i.e., integrals of Eq.2.1 over frequency and time delay, respectively) are equal to zero [43]. The time marginal indicates whether spectral filtering has occurred in the process of recording the FRPP trace, while the frequency marginal is an extremely sensitive tool to verify the instantaneous nature of the nonlinearity.

Typical FROG retrieval algorithms utilize the amplitude of the frequency-resolved signal measured in conventional FROG schemes [38]. However, due to heterodyning, the real part of the signal is detected in the FRPP method (Eq.2.1). Therefore, we had to develop a specialized algorithm aimed at obtaining the best match for the measured and computed FRPP matrices.
The *rms* difference between the two traces is minimized by a standard Levenberg-Marquardt least-squares fit routine [52]. A successful algorithm convergence results in a complete amplitude and phase reconstruction of the pulse. To accelerate the pulse retrieval, the spectral amplitude and phase of the pulse are represented by a cubic spline drawn through a number of points (nodes) that are equidistantly spaced within the frequency interval of interest. The influence of each node is restricted to the two adjacent spline segments on each side providing the required continuity of spectral phase and amplitude. The pulse reconstruction starts with a modest amount of nodes (typically 14) that are assigned random initial values. At this stage, the algorithm captures the most essential overall features of the pulse. As the algorithm begins to stagnate because of the coarse sampling, the amount of the nodes is automatically incremented. This provides a self-adjustment to any conceivable pulse complexity and substantially reduces the retrieval time. Fig.2.6 shows the reconstruction results (solid dots) along with final number of nodes and computational times. Typically, the calculation time does not exceed several seconds on a moderate Pentium-III 1-GHz PC. Note that the proposed algorithm does not require the pulse spectrum as additional input. Therefore, the latter can be used along with marginals as yet another independent test of the retrieval quality.

Figure 2.7a depicts the experimental FRPP trace of IR pulses obtained using a 2 mm thick CaF$_2$ sample. A nitrogen-cooled InSb photodiode (Hamamatsu P5968-200) and a lock-in amplifier (Stanford Research Sys.) were used to detect the FRPP signal after a scanning monochromator (CVI). It took ~15 min to acquire a typical trace. No appreciable (>1%) Raman contribution to the signal was revealed, justifying the assumption of the nonlinearity as instantaneous. The frequency-integrated SD signal amounted to only ~10% of the FRPP one putting SD FROG far beyond detection capabilities. A direct comparison of Fig.2.7 with Fig.2.6 immediately indicates that the IR pulse carries a predominantly quadratic spectral phase. As can be judged from Fig.2.7b, the reconstructed FRPP trace reproduces the essential features of the measured pattern fairly well. The intensity and phase of the IR pulses are given in Fig.2.7c (frequency domain) and Fig.2.7d (time domain). The pulse spectrum derived upon FRPP pattern reconstruction matches perfectly the experimental one (Fig.2.7c, solid dots) which demonstrates the high validity of the recorded FRPP trace. The pulse duration is ~70 fs while the spectral-limited value amounts to ~50 fs. The quadratic spectral phase that is already evident from Fig.2.7a, originates mostly from a GaAs substrate inserted in the IR beam, which serves as low pass filter.

### 2.4 Conclusions

We have constructed an optical parametric amplifier system for the generation of mid-IR laser pulses, tunable in the range of 2800-3800 cm$^{-1}$ with sub-100 fs duration and ~15 μJ energy. These characteristics match the requirements for laser pulses appropriate for nonlinear spectroscopy on OH-stretch vibration of water molecules.

A novel method was proposed for characterization of ultrafast IR pulses. It combines the high reliability inherent to FROG with an experimental simplicity keenly desirable in the IR. We have demonstrated that a standard pump–probe setup can be readily converted for pulse characterization by simply replacing the sample with a piece of a material transparent in the IR. Furthermore, for a vast majority of experiments in the liquid phase, FRPP pulse characterization can be conducted on a pure solvent with fast electronic response prior to introducing the sample molecules into it. We believe that features of FRPP as intrinsic phase-matching and great simplicity make it superior to any other reported method for characterization of fs optical pulses down to 1 fs [53].
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References


