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Adiabatic frequency conversion of ultrafast pulses

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Abstract A new method for efficient, broadband sum and difference frequency generation of ultrafast pulses is demonstrated. The principles of the method follow from an analogy between frequency conversion and coherent optical excitation of a two-level system. For conversion of ultrafast pulses, the concepts of adiabatic conversion are developed further in order to account for dispersion and group velocity mismatch. The scheme was implemented using aperiodically poled nonlinear crystals and a single step nonlinear mixing process, leading to conversion of near-IR (∼790 nm) ultrafast pulses into the blue (∼450 nm) and mid-IR (∼3.15 μm) spectral regions. Conversion bandwidths up to 15 THz FWHM and efficiencies up to 50% are reported.

1 Introduction

Nonlinear frequency conversion can be used to generate light in wavelength regions that are not easily accessible with commonly available laser systems. However, in most practical implementations, there is a trade-off between the bandwidth and efficiency of the conversion [1, 2]. In particular, for conversion of ultrafast light sources, most approaches address either the efficiency or bandwidth of the process, but usually not both [3, 4]. Commonly used ultrafast lasers such as Ti:Sapphire (Ti:S) produce very wide bandwidths but are limited in their tunability. Wavelength conversion of ultrafast pulses is straightforwardly done via frequency doubling [5, 6] or using an optical parametric amplifier (OPA) [7], but in order to preserve the wide bandwidth of the pulses and avoid dispersive effects, one needs to use relatively short nonlinear crystals. Thus, to retain the large bandwidth and the spectral phase characteristics of the pulse, one must settle for using shorter nonlinear crystals and relatively low conversion efficiencies.

In most methods of frequency conversion, it is essential to maintain small phase mismatch Δk between the interacting beams. In these cases, the conversion efficiency often remains high only for a narrow region of bandwidth. Adiabatic frequency conversion circumvents this trade-off and achieves conversion of broad bandwidth pulses with very high efficiency. The method was first realized for sum frequency generation (SFG) of narrowband pulses from the near-IR into the visible [8]. It was confirmed that the conversion process is insensitive to small changes in parameters that affect the phase mismatch such as temperature, crystal length, acceptance angle, and input wavelength [9]. Here, we generalize the method toward broadband ultrafast pulses and demonstrate efficient sum frequency generation (SFG) and difference frequency generation (DFG). This makes adiabatic frequency conversion an extremely flexible method for generating intense ultrafast pulses over a wide wavelength range covering the visible through to the mid-infrared.

The paper is organized as follows. In Sect. 2, we review the basic concepts of adiabatic frequency conversion.

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In Sect. 3, we develop the theoretical concepts that are necessary for performing adiabatic conversion in the ultrafast regime. The design of the experiments is described in Sect. 4, and we present the experimental results in Sect. 5. A discussion of the results, including future prospects and potential applications, is outlined in Sect. 6.

2 Principles of adiabatic conversion

The processes of SFG and DFG in the undepleted pump approximation were shown to be analogous to a two level atomic system induced by coherent light [8, 9]. Our understanding of these analogous systems can be used for formulating new frequency conversion schemes. Here, we consider the use of established adiabatic techniques for performing efficient population transfer in multilevel atomic and molecular systems [10, 11], and adapt them to the context of frequency conversion using broadband, ultrafast pulses. Adiabatic schemes in optics tend to be particularly attractive because nonlinear optical phenomena can remain robust to small changes in parameters that affect the phase evolution of the process [12].

It follows from this analogy that in order to produce an adiabatic passage of energy from the seed beam to the frequency converted beam, the phase mismatch parameter should vary slowly during the conversion process, from a large negative phase mismatch value to a large positive one (or vice versa). The phase mismatch Δk between the seed (ω_1), pump (ω_2) and SFG ($\omega_3 = \omega_1 + \omega_2$) or DFG ($\omega_3 = \omega_1 - \omega_2$) beams must be swept from $\Delta k < 0$ to $\Delta k > 0$ along the propagation axis in a nonlinear crystal. In the presence of a sufficiently strong pump field ω_2, energy is efficiently converted from frequency ω_1 to ω_3, analogous to adiabatic population transfer in two-level quantum systems. Thus, our method relaxes the restriction of minimizing the phase mismatch between the interacting beams, and instead requires Δk to be varied slowly (i.e., adiabatically) during the nonlinear interaction. The adiabaticity condition can be written:

$$\frac{d\Delta k}{dz} \ll \frac{(\Delta k^2 + \kappa^2)^{3/2}}{\kappa}, \tag{1}$$

where $\Delta k = k_1 + k_2 - k_3$ is the phase mismatch, where k_1, k_2, and k_3 are the wavenumbers of the seed, pump and frequency converted (either SFG or DFG) waves respectively; z is the position along the propagation axis, and the coupling coefficient $\kappa = \frac{4\pi\omega_1\omega_3}{\sqrt{k_1 k_3}c^2}\chi^{(2)}A_2$, where ω_1 and ω_3 are the frequencies of the seed and frequency converted waves, c is the speed of light in vacuum, A_2 is the pump amplitude and $\chi^{(2)}$ is the second-order susceptibility of the crystal.

With quasiphase matching (QPM), it is possible to tune the spatial structure of the poling domains in a nonlinear crystal and to construct almost any desired function of the phase mismatch parameter. A straightforward way to realize the adiabatic condition of (1) experimentally is to use a linear chirp poling, in which $d\Delta k/dz$ is linear in z. This simple type of aperiodic domain structure has been shown to be practical in a number of applications in laser engineering and ultrafast optics [13–16].

3 Frequency conversion of ultrafast pulses

For frequency conversion of quasi-monochromatic laser beams, one can omit the influence of the waves' group velocities, however, for ultrafast pulses it is essential to account for the dispersion properties of the nonlinear material. Material dispersion causes temporal spreading during propagation. Group Velocity Mismatch (GVM) results in temporal walk-off between the incoming seed pulse and the generated sum frequency pulse, which propagate in the crystal at group velocities v_{g_1} and v_{g_3}, respectively. The walk-off limits the useful nonlinear interaction length between the pulses, and is typically characterized by the quasistatic interaction length $L_{\mathrm{QS}} = \tau/GVM$, where τ is the temporal width of the seed pulse and GVM is the difference between the inverse group velocities of the pulses, defined as $\frac{1}{v_{g_1}} - \frac{1}{v_{g_3}}$. We also define the adiabatic effective length, $L_{\mathrm{adiabatic}} = \frac{\kappa}{d\Delta k/dz}$, where κ is the coupling coefficient between ω_1 and ω_3, and $d\Delta k/dz$ is the sweep rate of the phase mismatch along the propagation axis in the crystal. The quantity $L_{\mathrm{adiabatic}}$ is the effective length over which the adiabatic frequency conversion process takes place within the nonlinear crystal, and its definition is an adaptation of the adiabatic transition time of a general two level quantum system [17, 18], applied to the context of frequency conversion. To overcome the problem of GVM, we require that $L_{\mathrm{QS}} > L_{\mathrm{adiabatic}}$, so that the temporal walk-off between the pulses over the adiabatic interaction region is minimal. This sets a limit for the minimum temporal width of the seed pulse, which must be stretched to a duration τ_{min} which we define as:

$$\tau_{\mathrm{min}} > \left| \frac{\kappa}{d\Delta k/dz} \left(\frac{1}{v_{g_1}} - \frac{1}{v_{g_3}} \right) \right|. \tag{2}$$

Calculated values of τ_{min} for several typical experimental realizations are shown in Table 1. In each case, the pump wavelength and intensity were chosen to be 1053 nm and 150 MW/cm^2. Additionally, the sweep rate $d\Delta k/dz$ was assumed to be linear in z, and was chosen such that the average conversion efficiency was ~90% for the given crystal parameters and laser wavelengths. The maximum wavelength range supported by an aperiodically poled nonlinear crystal is given by the approximate formula

Table 1 Calculated values of $L_{\text{adiabatic}}$, the maximum bandwidth of the ω_1 seed beam, and $\tau_{\min}$ for various typical experimental realizations. See text for details

Nonlinear process	Seed (ω_1) central wavelength (nm)	Output (ω_3) central wavelength (nm)	Crystal type	Sweep rate ($1/\text{cm}^2$)	Crystal length (mm)	$L_{\text{adiabatic}}$ (mm)	Maximum seed bandwidth (nm)	$\tau_{\min}$
SFG	790	450	KTP	350	30	0.7	70	0.61
	790	450	SLT	250	30	0.8	55	0.72
	1550	630	KTP	120	20	0.9	130	0.38
	1550	630	SLT	90	20	1.2	80	0.57
DFG	790	3200	KTP	50	20	1.6	55	0.4
	790	3200	SLT	40	20	1.8	35	0.43

$(c/n_1(\omega_1))(d\Delta k/dz)L$, where c is the speed of light in vacuum, $n_1(\omega_1)$ is the refractive index of the central frequency of the seed inside the crystal, and L is the crystal length. Since the crystal length L is much larger than $L_{\text{adiabatic}}$, the bulk of the conversion takes place in a relatively small region of space in the center region of the crystal. Typically, a stretched pulse of duration 1–10 ps, tending toward the lower part of that range, is sufficient for conversion of femtosecond pulses in the visible and near-to-mid infrared spectral range.

Note that the converted pulse must then be recompressed after exiting the nonlinear crystal in order to recover the bandwidth-limited pulse duration. However, if the seed pulses are produced by a chirped pulse amplifier then it may be possible to bypass the compressor in that system and use the uncompressed pulses for adiabatic frequency conversion, provided that the uncompressed amplified pulses have pulse durations longer than the required $\tau_{\min}$. Since typical outputs of chirped pulse amplifiers are in the range of a few picoseconds to a few hundred picoseconds, this requirement can almost always be met quite easily. Thus, one can avoid incurring added stretcher and compressor losses on the seed pulses before performing the adiabatic conversion, thereby increasing the effective optical-to-optical efficiency of the conversion while at the same time reducing the complexity of the setup.

4 Experimental methods

The implementation of the adiabatic frequency conversion scheme is shown in Fig. 1. With the exception of the compressor stage, which has not yet been built into the experiment, we used the same setup for both the SFG and DFG implementations. The pump pulses, with energy ~1 mJ and duration 100 ns, were produced by a 1 kHz Q-switched Nd:YLF laser at 1053 nm and electronically synchronized in time to the output of a Ti:S multipass amplifier (Femtolasers GmbH) that was used as a source for the seed pulses. The seed pulse spectrum was centered near 790 nm with approximately 30 nm FWHM bandwidth and a transform limited pulse duration of 30 fs. We typically used seed pulse energies of 0.5–1.5 μJ, and either stretched these pulses in glass to a duration of 1–3 ps or alternatively used the uncompressed output of the Ti:S amplifier. This satisfies the requirement set by (2) and reduces unwanted second harmonic generation of the seed that would lower the efficiency of the conversion process.

For SFG, we designed an aperiodically poled KTP crystal in which the periodicity was linearly varied from 5.16 to 5.58 μm along a crystal length of 3 cm. This design enables conversion of 80 nm of total bandwidth from seed pulses centered at 790 nm. For DFG, we designed an SLT crystal in which the periodicity was linearly varied from 22.6 to 23.4 μm along a crystal length of 2 cm, which enables conversion of 20 nm of bandwidth from the near-IR seed pulses into the mid-IR. Both aperiodically poled crystals were uncoated. The seed and pump beams were focused to beam diameters of 100 μm at the centers of the crystals using spherical lenses with focal lengths of $f_1 = 70$ cm and $f_2 = 50$ cm, respectively. The pump intensity at the focus was 50 MW/cm^2. The beams are separated after the crystal using dichroic filters and the spectra of the seed and frequency converted beams were separately measured in a spectrometer (Jobin Yvon Triax 320).

5 Results

The spectra of the frequency converted pulses and their corresponding efficiency curves are shown for SFG in Fig. 2, and for DFG in Fig. 3. The efficiency curves depict the fraction of seed energy that is converted as a function of seed wavelength. Absorption losses in the crystals are negligible at the seed wavelength, so in calculating the efficiency we can assume that all the depleted energy contributes to frequency conversion. Although we have not yet performed the post-compression and characterization of the pulses, we have observed excellent conversion of the spectral shape and

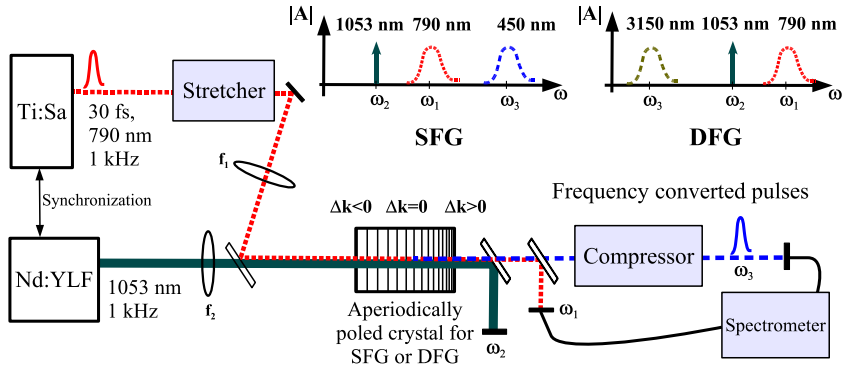

Fig. 1 Implementation of ultrafast adiabatic frequency conversion. The stretched seed pulse from a Ti:S multipass amplifier (*red dotted line*) and a strong pump beam from a Q-switched Nd:YLF laser (*dark green solid line*) are mixed in the adiabatic aperiodically poled nonlinear crystal. The frequency converted beam (*long blue dashed line*) is separated from the seed beam using a dichroic filter and their spectra are recorded separately in a spectrometer

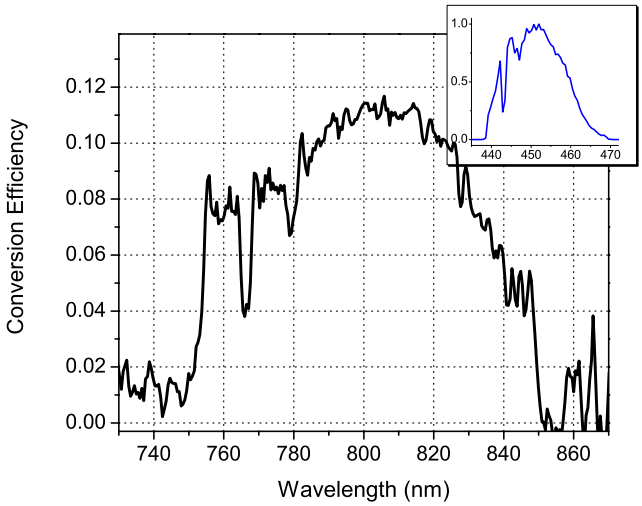

Fig. 2 Conversion efficiency of the seed pulse for ultrafast adiabatic SFG. The efficiency curve is nearly flat over the bandwidth of the seed pulse, indicative of the robustness of the adiabatic frequency conversion method. *Inset*: normalized spectrum of the SFG pulse as a function of wavelength in nm

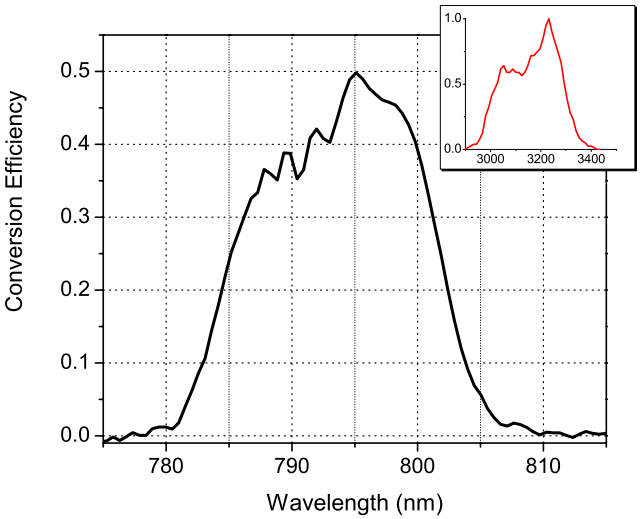

Fig. 3 Conversion efficiency of the seed pulse for ultrafast adiabatic DFG. The peak efficiency reaches 50%, and remains high (≥35–40%) over 20 nm of seed bandwidth. *Inset*: normalized spectrum of the DFG pulse as a function of wavelength in nm

bandwidth of the seed to both the visible and mid-IR wavelength regions.

5.1 Sum frequency generation—conversion from near-IR to visible

As shown in Fig. 2, the spectrum of the blue SFG pulse is centered near 450 nm and its bandwidth is sufficient for producing 30 fs pulses if compressed to the transform limit—a pulse duration that would be equal to that of the fully compressed seed beam. The efficiency curve is nearly flat across the seed bandwidth (falling off only in the tails of the spectrum), indicating the robustness of the adiabatic frequency conversion method. The dip in the efficiency curve near 765 nm is the result of a manufacturing defect in the crystal, which resulted in reduced conversion efficiency in a small portion of the seed spectrum.

5.2 Difference frequency generation—conversion from near-IR to mid-IR

The mid-IR downconverted spectrum is shown in Fig. 3 and is centered near 3150 nm, while the efficiency curve is flat over the bluer wavelengths and peaks slightly near the cut-off wavelengths further toward the red. Owing to the crystal design, nearly two-thirds of the 30 nm FWHM seed bandwidth was converted to the mid-IR, but the complete seed bandwidth could be converted by designing a longer aperiodically poled crystal or by altering the spatial sweep rate of the poling period.

5.3 Tunability of the frequency converted output pulses

The tunability of the center wavelength of the converted beam is dependent on the bandwidth of the seed and the

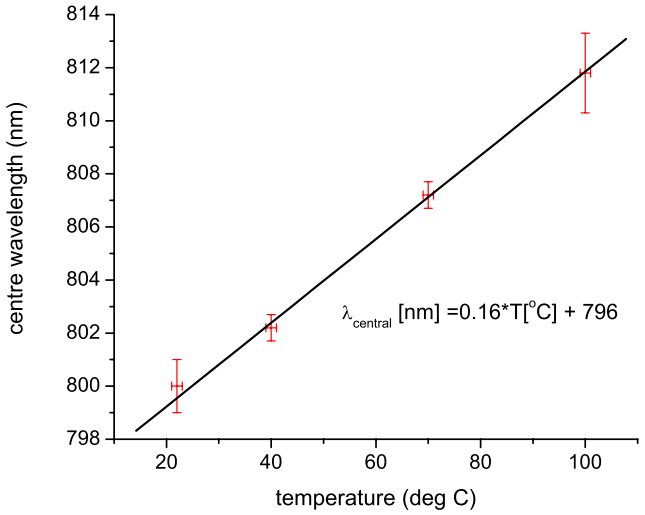

Fig. 4 Temperature tuning of adiabatic DFG. By tuning the temperature of the adiabatic crystal over an 80°C range, the seed depletion spectrum shifts by ∼12 nm, corresponding to more than 200 nm of central wavelength tunability in the mid-IR

tunability of the center wavelength of the pump. Since wavelength tuning of the Q-switched pump tends to be difficult, we chose to tune the converted DFG wavelength by changing the temperature of the adiabatic nonlinear crystal. The temperature-dependent change in the refractive index of the QPM crystal alters the magnitude of the phase mismatch Δk in each poling period, which in turn affects the peak of the seed beam depletion spectrum and the center wavelength of the frequency converted beam. The observed shift of the centroid of the seed depletion spectrum is shown in Fig. 4. The data shows a clear linear dependence of the shift with respect to the crystal temperature, and the observed shift of 0.16 ± 0.02 nm/°C is in good agreement with the calculated shift of 0.13 nm/°C according to the temperature-dependent Sellmeier equations for SLT crystals [19]. Thus, over the 80°C range of temperatures used in these measurements, the ∼12 nm shift of the center of the seed depletion spectrum is equivalent to more than 200 nm of tunability in the mid-IR.

6 Discussion

The conversion efficiency reaches a peak of ∼11% for SFG and 50% for DFG. Both these numbers are in good agreement with the expected efficiencies [9]. Losses due to possible fabrication errors in the crystal can therefore be assumed to be minimal. The maximum measured efficiency was limited in large part by the available pump power. Much higher efficiencies for both SFG and DFG, even approaching 100% in principle, are achievable by a sufficient increase in the pump intensity.

Adiabatic conversion using poled nonlinear materials offers many advantages compares to frequency conversion using birefringently phase matched crystals. Because of GVM, generation of sub-30 fs UV or visible pulses typically requires very thin crystals that limit the conversion efficiency [20, 21]. With birefringent crystals, the bandwidth is determined by the phase matching conditions in the crystal, which in turn is limited by its fundamental material properties. With adiabatic frequency conversion, the bandwidth is controlled by the length of the crystal and the sweep rate of Δk, both of which can be tailored. Provided that (2) for τ_{MIN} is satisfied, the conversion efficiency is determined solely by the adiabatic condition of (1), and is not influenced by the GVM in the crystal—in stark contrast to conversion using birefringently phase matched crystals.

Additionally, most ultrafast mid-IR pulse generation schemes involve visible or near-IR lasers that are converted to the mid-IR using multiple nonlinear conversion steps. For example, in a BBO-based OPA pumped by an 800 nm Ti:S laser, direct conversion to wavelengths greater than 2.5 μm is not practical because of poor gain efficiency and high GVM between the pump and signal or idler beams. Thus, one typically generates mid-IR pulses with Ti:S via a cascaded scheme involving an OPA followed by DFG between the signal and idler beams of the OPA [7]. With two nonlinear conversion steps, one pays a heavy cost in overall efficiency. In contrast, our scheme enables scalable near-IR to mid-IR ultrafast conversion with a *single* step.

Adiabatic frequency conversion appears especially promising for conversion of weak, nJ-level ultrafast pulses or broadband continuum pulses, where the intensity is too low for conversion via processes such as SHG. Even in situations where some conversion efficiency is feasible, such as cross-correlation upconversion between a nJ-level pulse and a much stronger pulse, one normally cannot afford low conversion efficiencies because of the already low signal levels. Moreover, it is expected that our method can be straightforwardly scaled to higher output powers, provided that the pump intensity does not exceed the damage threshold of the nonlinear crystal, and is strong enough to maintain the adiabaticity of the conversion. The fluence of the seed beam (and thus, the frequency converted output) can be increased as long as the pump remains undepleted. This may require a corresponding increase in the pump power as well as an increase in the spot sizes of both beams in order to avoid crystal damage. Alternatively, one can improve the damage threshold of the crystals by using shorter pump pulses. For the ∼100 ns pump pulses that were used in the experiments presented here, the damage threshold is approximately 150 mW/cm^2 for the aperiodically poled KTP crystal, and 300 mW/cm^2 for the SLT crystal. However, for ∼1 ns pulses, the damage thresholds are expected to be one order of magnitude higher.

Another important advantage with this method is that the spectral phase of the ultrafast pulse is preserved in the frequency converted field. This has important implications for

generating shaped ultrafast pulses in wavelength regimes where direct shaping is difficult or inaccessible using currently available pulse shaping technology.

To summarize, we have applied the adiabatic frequency conversion method for the conversion of ultrafast pulses for the first time. We show SFG and DFG conversion of near-IR ultrafast pulses to the visible and mid-IR spectral ranges. KTP and SLT nonlinear optical crystals with chirped poling periods were used to implement the conversion. We observe peak efficiencies of 50% for conversion of near-IR pulses from a Ti:S laser directly into the mid-IR in a single DFG conversion step. We predict much higher efficiencies for both SFG and DFG using more powerful pump lasers. The converted output can also be tuned by changing the temperature of the poled nonlinear crystals. We expect that this scheme can be scaled up in energy and can simplify ultrafast pulse shaping over a wide range of wavelength regimes.

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