Generation of narrow-bandwidth tunable picosecond pulses by difference-frequency mixing of stretched pulses

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We propose what is, to our knowledge, a novel method of generating tunable narrow-bandwidth picosecond pulses in which the chirp of equally stretched pump and signal is canceled by difference-frequency mixing. The method is proved experimentally by generation of pulses of 3.3-cm⁻¹ bandwidth and 10-ps duration in a type I β barium borate (BBO) crystal, starting from 1-ps pulses. © 1999 Optical Society of America [S0740-3224(99)00409-9]

OCIS codes: 190.4970, 320.1590, 320.5540, 320.7110.

1. INTRODUCTION

Over the past decade, Ti:sapphire laser systems employing a chirped pulse amplification technique have become widely used in spectroscopic research. These systems generate energetic optical pulses whose duration is from tens of femtoseconds to a few picoseconds. However, numerous applications, especially in the IR, require nearly transform-limited pulses in the duration range of 2–20 ps, where application of chirped pulse amplification becomes problematic owing to the lack of bandwidth for efficient pulse stretching. Therefore nonlinear-optics-based methods that allow one to reduce the bandwidth without sacrificing efficiency are of particular interest.

Recently, a method for bandwidth narrowing was demonstrated in which the second harmonic or the sum frequency is produced by counterchirped pulses. The method allows comparatively easy adjustment of pulse duration, as well as wavelength tuning, within the bandwidth of the incoming pulses. The basic principles of the technique were shown to work approximately two decades ago, in Ref. 4, where chirp removal from randomly phase-modulated conjugated pulses was demonstrated. Extension of three-wave interactions to cascaded processes provided a tool for a more-complex combination of pulse chirps by sum-frequency generation and parametric amplification.

As for the IR range, tunable picosecond pulses have been obtained by use of optical parametric oscillators or amplifiers or by difference-frequency (DF) mixing between pulses from various laser sources: Nd-doped laser pulses and dye laser pulses, signal and idler pulses from an optical parametric amplifier, Nd:YAG laser pulses and the idler output of an optical parametric amplifier. Typical values of the time–bandwidth product for systems based on IR optical parametric amplification range from nearly transform limited to more than ten times transform limited, whereas systems employing DF mixing approach the Fourier transform limit.

In this paper we report a method of narrow-bandwidth pulse generation through DF mixing of stretched pulses of ∼1-ps initial duration. In a theoretical consideration we show that the group-velocity (GV) mismatch between pump and signal pulses is the main factor limiting the parametric crystal length suitable for narrow-band pulse generation. Experimentally, we demonstrate the generation of 10-ps, 3.3-cm⁻¹ pulses in a type I β barium borate (BBO) crystal.

2. THEORY

In this section the theoretical background of DF generation in a nonlinear crystal by frequency-chirped pulses is presented. By DF generation we mean the parametric amplification process, in which we have two pulses: a signal (seed) pulse, and a pump pulse injected into the crystal. While propagating through the crystal, the signal pulse is amplified by the pump pulse, and at the same time a DF pulse is generated.

According to the approach developed for broadband sum-frequency generation by phase-modulated input fields, we can characterize chirped pulses by defining an instantaneous frequency ω(t). In the case of DF mixing at the entrance of the crystal we have two phase-modulated pulses, whose frequencies vary in time as

\[ \omega_j(t) = \omega_{j0} + \beta_j t + \ldots, \]  

where the subscript j refers to the signal (s) or pump (p) pulse, \( \omega_{j0} \) is the central frequency, and \( \beta_j = \frac{d\omega_j}{dt} \) is the linear chirp. Such a notation is meaningful when one is using strongly chirped pulses. Below we restrict our analysis to the case of quadratic phase modulation or linear chirp of input pulses.

The pump and signal pulses with linear chirps generate the DF pulse with an instantaneous frequency that may be expressed according to the energy conservation law as

\[ \omega_{df}(t) = \omega_{df0} + \beta_p t - \beta_s(t + dt), \]
where \( \omega_{df0} = \omega_{p} - \omega_{s} \) and \( dt \) is the time delay between the pump and the signal pulses. From Eq. (2) it follows that, when the pump and signal chirps are set to be equal at the entrance of the nonlinear crystal, generation of a chirp-free DF pulse with the bandwidth limited only by the pulse duration takes place. This means that, if we stretch initial pulses 10 times, we can expect 10-fold bandwidth narrowing. In a real situation the narrowing is smaller because of the pulse-shortening phenomenon during parametric amplification.

The bandwidth reduction factor may also be diminished by GV mismatch unless some restrictions on the length of the crystal are established. The influence of GV mismatch can be understood as follows. When pump and signal pulses are matched, i.e., \( \beta_{p} = \beta_{s} = \beta \). However, a certain amount of time delay between the pump and the signal pulses is introduced while the pulses propagate through the crystal, owing to the GV mismatch. This causes a change in the DF at the end of the crystal by \( \beta L v_{ps} \), where \( L \) is the crystal length and \( v_{ps} = 1/u_{p} - 1/u_{s} \) is the (reciprocal) GV mismatch. It is reasonable to restrict the shift of the DF so that it is not larger than the DF bandwidth \( \Delta \omega_{DF} \), which gives the upper limit to the crystal length:

\[
L < \frac{\Delta \omega_{DF}}{\beta v_{ps}}. \tag{3}
\]

We can express the DF bandwidth through the duration \( \tau_{0} \) corresponding to the Fourier transform of the pump pulse spectrum as \( \Delta \omega_{DF} \approx \tau_{0} \beta \), not taking into account DF pulse parametric shortening, which makes inequality (3) even more pronounced. Under such an assumption inequality (3) changes to

\[
L < \frac{\tau_{0}}{v_{ps}} = L_{\text{walk-off}}, \tag{3'}
\]

where \( L_{\text{walk-off}} \) is the walk-off length. This parameter differs from that defined in conventional parametric interaction. The difference is in the pulse duration: We take the duration corresponding to the Fourier transform of the spectrum rather than the real duration of the interacting pulses. Hence the relevant parameter in the chirped pulse parametric amplification is the pulse bandwidth rather than its duration.

Besides the walk-off length, there is another important parameter in parametric amplification, namely, the gain bandwidth, which in ordinary parametric interaction is defined as

\[
\Delta \omega_{a} = \frac{4 \sqrt{\ln 2}}{v_{ps \| LL} \sqrt{L n}}, \tag{4}
\]

where \( v_{ps \| LL} \) is the GV mismatch between the signal and the DF waves and \( L _ {n} \) is the nonlinear length (\( L _ {n} = L 'n \), where \( L ' \) is a small-signal gain coefficient).

In the case of chirped pulses, assuming that the condition of perfect phase matching is met for the central frequencies \( \omega_{p0}, \omega_{s0}, \) and \( \omega_{DF0} (t = 0) \) and introducing a shift in time \( \delta t \), we get a shift in the pump frequency by \( \delta \omega_{p} = \beta \delta t \), which causes the signal gain band to slide along a frequency scale by \( \delta \omega_{a} = k \beta \delta t \), where \( k = v_{ps \| LL} / v_{ps \| LL} \). This modifies the signal gain bandwidth to

\[
\Delta \omega_{a} = \frac{\Delta \omega_{a0}}{|k - 1|} = \frac{4 \sqrt{\ln 2}}{|v_{ps \| LL} \sqrt{L n}|}, \tag{5}
\]

Thus we get the GV mismatch between the signal and the pump waves in Eq. (5) instead of the GV mismatch between the signal and the DF waves [Eq. (4)]. Inasmuch as the signal pulse is chirped, decrease in the gain bandwidth shortens duration of both signal and DF pulses and increases the DF bandwidth. We may avoid this situation by setting one more restriction on the crystal length:

\[
L < \left( \frac{4 \sqrt{\ln 2}}{\Delta \omega_{a} v_{ps \| LL} \sqrt{L n}} \right)^{2} = \left( \frac{\tau_{0}}{v_{ps \| LL} \sqrt{L n}} \right)^{2} \approx \frac{L_{\text{walk-off}}}{L_{n}}. \tag{6}
\]

The right-hand side of inequality (6) is reorganized by substitution of \( \Delta \omega_{a} \) with \( \Delta \omega_{p} \), since only those spectral components of the signal that lie under the pump may be amplified. Inequality (3') is stronger than inequality (6) if \( L_{\text{walk-off}} > L_{n} \). In fact, only this case is of interest to us here, since we can take \( L > L_{n} \), which allows us to obtain considerable amplification.

It is worth noting that, in the case of narrowbandwidth pulse generation by frequency doubling of chirped pulses in a type I crystal \(^{2}\) there is no GV mismatch between the generating pulses, since both pulses are of the same wavelength and polarization. Hence a crystal as thick as is required for efficient conversion may be used without any negative influence on the generated second-harmonic bandwidth. Unfortunately, this is not the case in the DF generation.

Figure 1 shows the values of \( L_{\text{walk-off}} \) for 1-ps pulses in type I (ordinary signal and DF; extraordinary pump) and type II (ordinary signal; extraordinary DF and pump) BBO and AgGaS\(_{2}\) crystals and in an \( xz \)-cut type II (extraordinary signal; ordinary DF and pump) KTP crystal for the pump wavelengths of Nd:glass and Ti:sapphire lasers as a function of DF wavelength. BBO is used to generate visible and near-IR radiation of as much as 2.7 \( \mu \)m, whereas, using KTP and especially AgGaS\(_{2}\), one can go...
farther into the IR region, to as high as 13 μm. The drawback of using AgGaS2 is that it cannot be pumped directly by a Ti:sapphire laser because of the two-photon absorption of 800 nm.16

An important parameter of any amplifier is the gain increment, which in our case may be defined as

\[
\text{Inc} = \ln \left( \frac{I_{\text{DF}}}{I_{\text{SO}}} \right),
\]

where \( I_{\text{DF}} \) is the output intensity of the DF wave (\( I_{\text{DF}} = 0 \) at the input, as stated in the beginning of this section) and \( I_{\text{SO}} \) is the input intensity of the signal wave. Neglecting the pump depletion and the GV mismatch, we can express the gain increment in the case of \( L \gg L_n \) as

\[
\text{Inc} = 2 \frac{L}{L_n} + \ln \left( \frac{1}{4Q} \right),
\]

where \( Q = \omega_{\text{DF}} / \omega_{\text{SO}} \) is the degeneracy parameter. Figure 2 shows gain increment values versus DF wavelength values for the same crystal and pump configurations as are shown in Fig. 1. The pump intensity in all the cases is 1 GW/cm², giving \( L_{\text{walk-off}} > L_n \) for all the crystals. The crystal lengths have been chosen to satisfy inequality (3') in the whole tuning range (we neglect any other effects, such as spatial walk-off of interacting beams or beam self-phase-modulation, which could limit the crystal lengths). Therefore, as shown above, inequality (6) is then automatically satisfied.

Inasmuch as type II BBO has longer \( L_{\text{walk-off}} \) than type I BBO, it supports a broader gain bandwidth even close to degeneracy, unlike in the traditional parametric amplification process. This result may be explained by the fact that the gain bandwidth in a type II BBO crystal slides synchronously with the signal frequency to be amplified. Hence this crystal is preferable when one is dealing with femtosecond pulses.

Contrary to the case of BBO, both types of interaction in AgGaS2 have similar values of walk-off length (Fig. 1). Still, there are some differences. On the one hand, the type II interaction has a narrower tuning range than the type I interaction, but, on the other hand, slightly higher amplification may be achieved in the type II crystal for wavelengths >3.5 μm (Fig. 2).

As can be seen from the same figure, AgGaS2 gives higher gain increment values than KTP for a 1055-nm pump, regardless of the former crystal's shorter applicable length, whereas 800-nm-pumped KTP seems to be most promising for wavelengths ranging to as much as 4.5 μm.

3. EXPERIMENT

In our experiment we used a Nd:glass laser system that delivered frequency-doubled 527-nm-wavelength pulses of 1.2-ps duration, 15-cm⁻¹ spectral bandwidth, and >3-mJ energy. The output of the laser was split into two branches that generated frequency-chirped pump and seed fields to be combined in a DF mixer (Fig. 3).

In the first branch the linearly chirped pump pulse was formed by use of a grating-pair pulse stretcher with a group delay dispersion (GDD) parameter of \(-8\) ps². At the output of the stretcher the pump pulses had a duration of \(\sim 18\) ps with a chirp parameter of \(\beta_p = -0.66\) cm⁻¹/ps.

In the second branch the seed pulses of tunable wavelength were created with an optical parametric generator–amplifier (OPG-A). The OPG-A was based on a type II BBO crystal seeded by spectrally filtered superfluorescent light. The OPG-A produced \(-0.9\)-ps, \(\sim 25\)-cm⁻¹ pulses within the 640–2600-nm-wavelength range. The idler wave of the OPG-A was directed to the second grating-pair stretcher such that both length and grating angle were adjustable, which made it possible to maintain constant GDD within the range of 1055–1090 nm. In such a way seed pulses of tunable wavelength and constant chirp were produced. In addition, by design, the stretcher was capable of sustaining a constant optical path length while being tuned.

The pulses stretched in both branches were retimed for optimal overlap and were combined in the 8-mm-thick type I BBO crystal. We used noncollinear geometry with an angle of 0.5 deg between the pump and the seed beams to be able to separate the generated DF wave. The pump beam with an energy of 1.1 mJ was telescoped in front of the crystal to a diameter of 2–2.5 mm, which corresponded to \(\sim 1\) GW/cm² pump intensity and 0.9-mm non-
linear length inside the crystal. As can be seen from Fig. 1, the walk-off length is >11 mm for ~1-ps pulses within the entire tuning range, which is longer than the crystal length and much longer than \( L_n \). Hence Eqs. (4) and (7) are more than satisfied, ensuring that GV mismatch effects will be negligible.

The parametric amplifier seeded by 2.5-\( \mu \)J pulses produced DF pulses with an energy of 150 \( \mu \)J, corresponding to 26% total conversion efficiency. We controlled the DF pulse bandwidth by tuning the length of the seed stretcher and thereby changing its GDD (Fig. 4). As can be seen, the DF bandwidth dependence has a well-expressed minimum point at which the linear chirps of pump and seed pulses compensate each other. The narrowest bandwidth achieved was 3.3 cm\(^{-1}\), which was almost eight times less than the initial bandwidth of the seed pulses (Fig. 5(a)). This bandwidth was sustained within the entire tuning range of 1025–1055 nm, which was limited only by the seed stretcher.

Figure 4 also displays the signal pulse bandwidth as a function of GDD. In contrast to the DF bandwidth, it monotonically shrinks as GDD increases. This result is quite obvious, considering that with an increase of pulse chirp the spectral components get farther apart from one another, and fewer of them fall under the pump pulse envelope.

We combined the DF beam and the OPG-A signal beam in a KDP crystal to measure DF pulse duration. Since the OPG-A signal pulse is more than ten times shorter than the DF pulse, DF pulse duration corresponds closely to the width of the cross-correlation function measured to be 10 ps [Fig. 5(b)]. This twofold shortening of the DF pulse as compared with the pump pulse (18 ps) is due to the Gaussian envelope of the gain profile. The time-bandwidth product of 10-ps DF pulses with a bandwidth of 3.3 cm\(^{-1}\) is more than two times larger than that of the transform-limited pulses. Nevertheless, it is much improved as compared with BBO-based OPG-A systems delivering a similar pulse duration and working at high conversion efficiencies.\(^7\)

4. CONCLUSIONS

In conclusion, we have demonstrated a method of narrow-bandwidth picosecond pulse generation in which difference-frequency mixing of chirped pulses is used. This method offers a convenient way of producing IR pulses in the duration range of 2–20 ps, with both wavelength and bandwidth being adjustable. We have shown that group-velocity mismatch between pump and signal pulses is the main factor limiting the parametric crystal length suitable for narrow-band pulse generation. Experimentally, 10-ps, 3.3-cm\(^{-1}\) DF pulses have been generated in a type I BBO crystal, starting from ~1-ps pulses.

ACKNOWLEDGMENTS

We thank G. Valiulis for providing software for our calculations and A. Piskarskas for providing a critical reading of the manuscript.

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