Generation of a high-energy ultrawideband chirped source in periodically poled $LiTaO_3$

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Received May 10, 2004; revised manuscript received October 27, 2004; accepted November 1, 2004

A method for generation of a chirped, ultrawideband infrared source by use of optical parametric generation in periodically poled crystals and pumped by a chirped Ti:sapphire laser is described. A ~35% bandwidth in the idler branch was demonstrated in a periodically poled $LiTaO_3$ crystal pumped by a chirped Ti:sapphire laser with 2.1% bandwidth. Optical parametric generation and optical parametric amplification configuration allowed us to generate up to a ~250-μJ chirped pulse from 2.1 to 3 μm. © 2005 Optical Society of America

OCIS codes: 190.0190, 190.4410.

1. INTRODUCTION

Generation of an ultrawideband chirped pulse is of great importance for time-resolved spectroscopy, remote sensing, process control, the generation of ultrashort pulses,[1,2] the controlled excitation of atoms and molecules by means of ladder-climbing[3–5] or autoresonance methods,[5,6] and the autoresonant excitation of a plasma beat-wave accelerator.[7] The ladder-climbing or autoresonance methods for the excitation of vibration levels in molecules requires a relatively high-energy, ultrawideband chirped pulse in the IR or mid-IR regime, since this is the range of molecular vibrations. The excitation of a plasma beat-wave accelerator by the method that was suggested by Lindberg *et al.*[7] also requires a relatively high-energy chirped pulse in the IR or mid-IR regime with a relatively long pulse duration (from few picoseconds to some hundreds of picoseconds).

The conventional method for generation of a chirped pulse is the stretching of a wideband, ultrashort pulse by means of dispersive apparatus.[2] The lack of an efficient wideband lasing material in the mid-IR and IR regimes prevents the use of this method directly. Another possibility is the use of a commercial ultrashort Ti:sapphire laser and the conversion of it by means of optical parametric generation (OPG) to the IR regime, followed by stretching. This method has several important drawbacks. First, there is usually a significant (tens of percent) loss of energy in the stretcher. Also, alignment of the stretcher in the IR is difficult, particularly for field applications. Finally, single-pulse energy and average power are limited by material damage constraints.

In this paper we propose an alternative method based on the stretched pulse of the Ti:sapphire laser, in which a significant amount of energy is available and stretcher alignment is not a problem. This chirped pulse at 0.8 μm is converted into a chirped pulse at the IR regime. This method has several advantages. First, no stretcher is needed in the IR, and thus there is no energy loss in the IR. Second, since the pump pulse is a relatively long chirped pulse and the damage threshold energy for a given crystal scales as the square root of the pulse duration, we can use more pump energy and get more energy in the IR. Also, the high pulse-repetition-rate capability of Ti:sapphire lasers and the favorable thermal properties of the crystal should enable high average power. Finally, the relative simplicity and ruggedness of the system should make it suitable for field or industrial applications, such as remote sensing and process control.

OPG, used in our scheme, is a nonlinear effect in which a high-intensity optical pump pulse produces in a nonlinear crystal two beams (signal and idler) that must satisfy the conditions $\omega_p = \omega_s + \omega_i$ (energy conservation) and $k_p = k_s + k_i$ (momentum conservation or phase matching). A chirped pump $[\omega_p = \omega_p(\mathrm{t})]$ will generate a chirped signal and idler pulses whose chirp bandwidth will depend on the dispersion of the nonlinear medium. If periodically poled crystals are chosen as the nonlinear medium, the (quasi) phase-matching relation is given by $k_p = k_s + k_i + 2\pi/\Lambda$ (Λ is the poling period) with phase-matching curves strongly dependent on Λ.[8,9] In our previous work[10] we measured the signal from a periodically poled KTP crystal with a 27.1-μm period, pumped by a Ti:sapphire laser. The signal from that crystal was positively chirped from $\lambda = 1.45\,\mu$m to $\lambda = 1.1\,\mu$m, corresponding to a bandwidth of 6.6×10^4 GHz.

2. EXPERIMENT

In this work we report on the OPG of an ultrawideband source in the range of 2.1–3 μm by use of a periodically poled stoichiometric $LiTaO_3$ (PPSLT) and the amplification of this radiation with a PPSLT optical parametric amplifier (OPA). Figure 1 illustrates the phase-matching curves of PPSLT held at a temperature of 150 °C, having a poling period of 22.8 μm and pumped by the Ti:sapphire laser. To calculate this curve we used the Sellmeier equation published by Bruner *et al.*[11] It can be seen that the 2.1%-wide Ti:sapphire-pump chirp can be converted to a ~35% chirp at the idler frequencies (2.1–3 μm). The PPSLT was chosen for this work owing to its high damage

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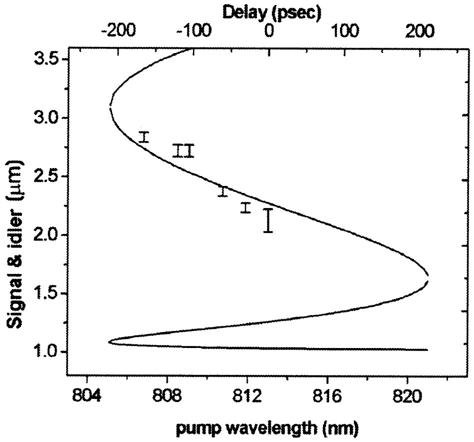

Fig. 1. Phase-matching curve of PP-SLT with a period of 22.8 μm, pumped around 810 nm. The bars on the graph represent cutoff wavelengths measured by the gated spectroscopy experiment (see below).

threshold and large effective nonlinear coefficient. During crystal preparation 1-mm-thick *z*-cut SLT wafers were sliced into ~9-mm-wide samples along the *y* crystallographic axis. Photoresist stripes were defined photolithographically on the z^+ surface of the sample and parallel to the *x* axis of the crystal. The poling was done in a vacuum chamber through application of high voltage pulses on the z^+ surface. After the poling was concluded the electrodes were removed and the end facets were polished.

Spectrum broadening due to noncolinear propagation is one of the main drawbacks of OPG configuration that allows usage of only a small portion of its energy. To overcome this energy limitation we took this portion and amplified it in a second-stage crystal. The combined OPG–OPA system consisting of a 0.5 mm × 8.5 mm × 23.5 mm PPSLT crystal serving as the OPG and a 1 mm × 10 mm × 18 mm PPSLT crystal serving as the OPA. To take advantage of the entire width of the OPA crystal, we focused the pumping Ti:sapphire laser into the crystal by a cylindrical telescope. A germanium window was used to separate the idler from the signal. The resultant idler energy was up to 250 μJ.

The major goal of this experiment was to test both the bandwidth and the chirp of the idler. Figure 2 shows the experiment setup. The PPSLT crystal was pumped by a Ti:sapphire laser with a chirped-pulse duration of ~350 ps FWHM and a bandwidth of 2.1% around 810 nm. The amplified stretched pulse was split into two beams, approximately 1 mJ of the stretched pulse, and was focused into a periodically poled $LiTaO_3$ crystal. The other part of the beam was compressed to an ~200 fs pulse and served as a probe. The spectrum of the idler signal generated by the PPSLT was analyzed with a 1-m McPherson (McPherson, Chelmsford, Massachusetts) scanning monochromator. The obtained spectra are shown in Fig. 3. The spectra exhibits, in addition to the broad IR emission, absorption features around 2.7 μm owing to the water vapors. Purging the system with argon flow eliminates this absorption band.

To obtain a temporal resolution of the spectrum we employed an optically gated frequency-resolved technique, resembling the FROG technique[12] but for subnanosecond pulses. A laser-induced electron-hole plasma in ZnSe window, placed in front of the spectrometer, served as the gating mechanism. The plasma reflected the IR radiation if its frequency was below the corresponding IR frequency. For this purpose, the probe pulse was focused on the ZnSe window to gate the delayed idler by forming dense plasma inside the ZnSe. By varying the delay line we could gate the idler at various times and therefore cut the spectrum at different places. Figure 4 shows the overlaid ungated and gated spectra at various delay times. It can be seen in each frame that, at the lower part of the spectrum, the ungated and the gated spectrum are essentially the same, but at some cutoff wavelength they depart from each other. This technique allowed us to measure the cutoff wavelength (defined at the point of separation between the gated and ungated spectra at a given delay) as a function of the delay and to derive the chirp function of the idler signal. Note there are few changes in the spectrum from one delay to the other.

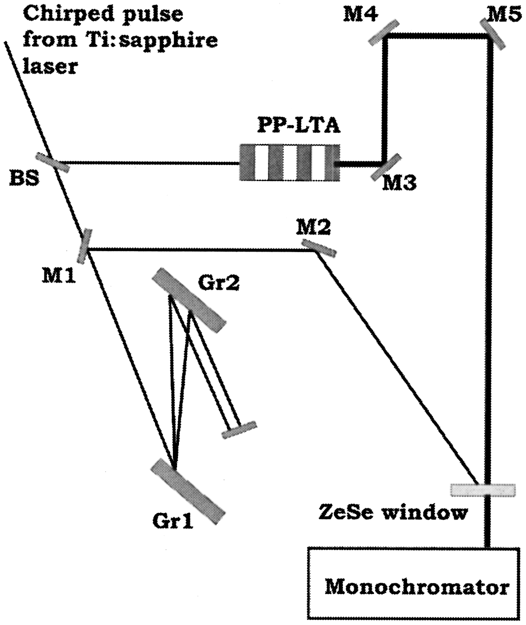

Fig. 2. Experiment setup. BS, Beam splitter; M, mirror; G1, G2, grating.

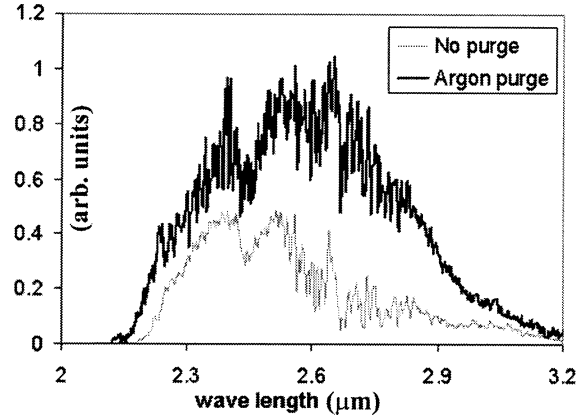

Fig. 3. Idler spectra purged with argon (black) and without purging (gray).

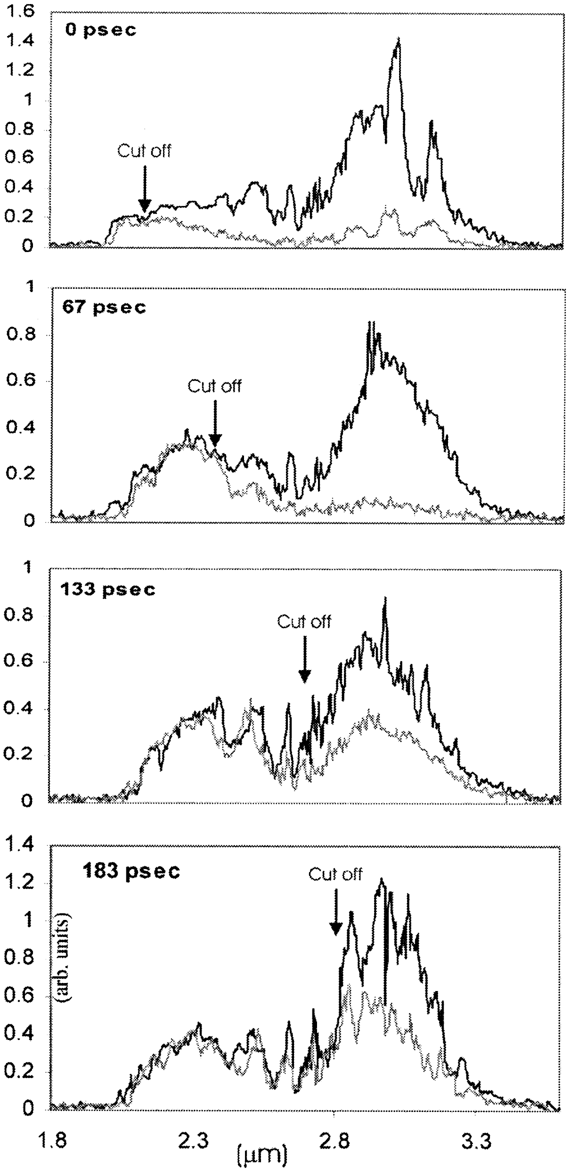

Fig. 4. Gated spectra (gray curve) versus ungated spectra at various delays.

These changes are due to the long scanning time of the spectrum and long term instabilities of the Ti:sapphire laser spectrum. All of the spectra in this figure were obtained without a purge of the system. The relatively high intensity around 2.9 μm is due to a spike in the Ti:sapphire laser spectrum.

The obtained broadband emission in range 2.1–3 μm exhibits an IR chirped pulse with a bandwidth of 35%. Using our knowledge of the chirp function of the pump, we could superimpose the cutoff wavelength versus the pump wavelength. The obtained results are presented by the bars in Fig. 1. The bar heights represent the accuracy in the definition of cut-off wavelength. Comparison between the theoretical curve and the experimental curve shows quite satisfactory agreement.

In conclusion, we have demonstrated generation of high-energy, ultrabroad ~35% chirped IR radiation in the 2.1–3 μm range using a PPSLT crystal pumped by a chirped 2.1% wide Ti:sapphire laser.

ACKNOWLEDGMENTS

The authors are very thankful to R. Hubburd from Naval Research Laboratory for his highly useful comments.

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