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Generation of ultrawide-band chirped sources in the infrared through parametric interactions in periodically poled crystals

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A method to generate chirped ultrawide-band sources with a chirp bandwidth of about 50% in the infrared is described and experimentally verified. It is based on optical parametric generation in periodically poled crystals with a chirped Ti:sapphire as a pump source. We have demonstrated a 27% wide bandwidth in the signal branch and 45% bandwidth in the idler branch when a periodically poled KTP crystal was pumped by a chirped Ti:sapphire laser with 12 nm full width at half maximum bandwidth. [DOI: 10.1063/1.1537871]

The ability to place an atom or a molecule in a specific state is of great importance in spectroscopy and chemical dynamics.[1] Direct excitation of the atom or the molecule with monochromatic radiation that matches the desired energy gap is one way to achieve this goal. However, excitation of highly excited vibrational states in molecules is inefficient, due to the small value of the transition dipole moment between the initial and final states.[2] Another option is to cause a cascaded transition from the initial to the final state through a series of intermediate levels, by a pulse with a continuously variable frequency. Such pulse is said to be chirped. Many authors address this problem, both classically and in quantum mechanical terms.[2–5] Recently, this method was demonstrated in atomic and molecular systems.[2,6] For example, Maas *et al.* used a chirped pulse at 5.4 μm with a bandwidth of $\Delta\omega/\omega=0.027$ to excite NO to its third vibrational level. In order to excite targets to higher levels, a chirped infrared radiation source with a wider bandwidth is desired. Chirped ultrawide-band infrared sources are also useful for application such as time-resolved spectroscopy and adaptive pulse compression.[7] Advances in electrical poling of ferroelectric, nonlinear materials have made periodically poled crystals such as $LiNbO_3$,[8,9] $KTiOPO_4$ (PPKTP),[10] and recently, stoichiometric $LiTaO_3$[11] available. They offer higher nonlinear coefficients, noncritical phase matching in their entire transparency range and wide phase-matching bandwidths. By a suitable choice of crystal, type of phase matching and poling period, one can obtain ultrawide-band pulses in any spectral range up to 5.5 μm. For example, a type I (*yyz*) parametric generator in PPKTP with a $\sim$7.4 μm poling period, allows to obtain a bandwidth of 1.3 μm around a central wavelength of 2.2 μm.

In this letter we demonstrate a PPKTP parametric generator with a chirp bandwidth of $\Delta\omega/\omega=0.27$ and $\Delta\omega/\omega=0.45$.

Optical parametric generation (OPG) 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 $[\omega_p=\omega_p(t)]$ will generate 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 $k_p=k_s+k_i+2\pi/\Lambda$[8,12] (Λ is the poling period) with phase matching curves strongly dependent on Λ. Figure 1 shows the calculated phase matching curve of a PPKTP crystal with a 27.1 μm period, pumped by a Ti:sapphire laser. This specific configuration allows for two separate solutions that satisfy the phase-matching conditions. A 2.1% wide Ti:sapphire-pump chirp is converted here to a $\sim$27% chirp at the signal frequency (1.1–1.45 μm).

Figure 2 shows the experimental arrangement for converting the chirp of a stretched Ti:sapphire laser to a chirped beam in the mid-infrared (IR) range. The pump laser is Ti:sapphire with a bandwidth of $\sim$10 nm full width at half maximum (FWHM). Central wavelength is tunable from 790 to 820 nm. The pulse is stretched to 400 ps FWHM and amplified up to 20 mJ. The laser was collimated to a 0.4 mm radius beam and passed through a 30 mm long, temperature controlled PPKTP crystal with a poling period of 27.1 μm. The crystal facets were uncoated. To prevent damage to the

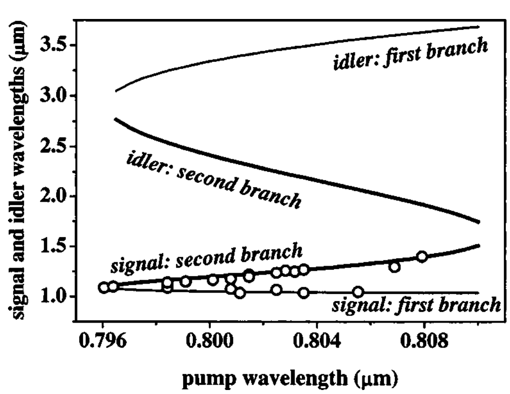

FIG. 1. Phase matching condition in PPKTP with period of 27.1 μm pumped by a $\sim$12 nm chirped Ti:sapphire laser. Open circles are experimental data. The curve was calculated from Sellmeier's coefficients in Ref. 10.

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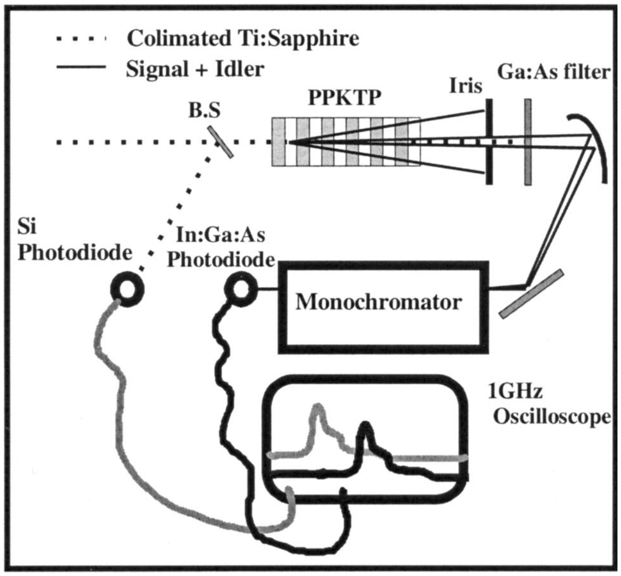

FIG. 2. Experiment setup. The spectrum is scanned by a monochromator and InGaAs photodiode. The chirp was measured by measuring the delay between the two photodiode signals, as a function of the wavelength.

crystal facets, care was taken not to exceed 2 GW/cm^2 in power density (~4 mJ). The pump, signal, and idler were polarized along the z axis of the crystal. The output radiation from the PPKTP was passed through an iris and a Ga:As window that served to block the 800 nm pump. The signal was focused into a monochromator and detected by a fast InGa:As photodiode.

The chirp bandwidth was determined by measuring the delay between the output from the Si photodiode that was placed before the PPKTP crystal and acted as a reference marker, and the output from the InGaAs photodiode placed behind the nonlinear crystal and the monochromator. For simplicity we choose to measure the spectrum of the signal branches only (up to 1.45 μm). Figure 3 shows the delay as a function of wavelength. The two signal lobes can be clearly recognized.

In order to fit the delay to the phase matching curve, another set of measurements was made to find the relation between the delay and the pump wavelength. This was done by creating a gap in the pump's spectrum, by covering a selected area of the stretcher's grating, and identifying the corresponding gap in the spectrum of the signal, as shown in

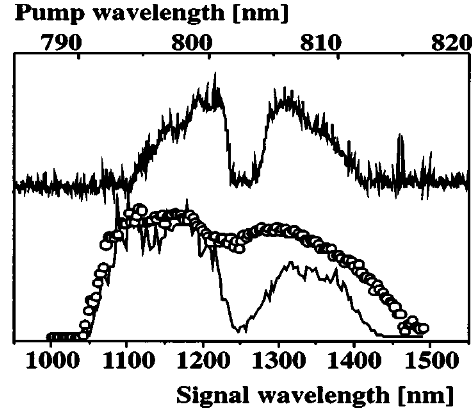

FIG. 4. Spectrum of the laser (upper trace) and signal (lower trace). Blocking some of the seeding spectrum in the regenerative amplifier creates the gap in the spectrum. The circles show the broadening of the spectrum due to noncollinear phase matching (taken without an iris). The continuous lower trace shows the signal spectrum when an iris blocks noncollinear beams. The intensities of the two spectra were rescaled.

Fig. 4. When the entire signal beam was focused into the monochromator, noncollinear phase matching masked the expected dip in the signal spectrum. This effect causes a wavelength shift in the signal as function of its propagation angle.

The correspondence between the signal delay and the originating pump wavelength was determined from the location of this gap, together with a separately measured delay curve of the pump wavelengths. The calculated results are superimposed as open circles on the phase-matching curve in Fig. 1. These results confirm that the two signal branches are chirped as expected. We have achieved a chirp of 27% in the upper lobe of the signal corresponding to a chirp of 45% in the corresponding idler beam. The conversion efficiency of the OPG was determined by inserting a filter that is opaque in the idler band and measuring the output energy as function of the pump energy (see Fig. 5). Taking into account losses due to filters and Fresnel reflections, a maximum of 45% signal conversion in both branches was obtained at 4.3 mJ, corresponding to approximately 70% in pump depletion. For practical purposes, however, the available energy is obviously lower since this value comprises also the noncollinearly matched frequencies.

A practical method to generate chirped ultra wide-band sources by optical parametric generation in periodically

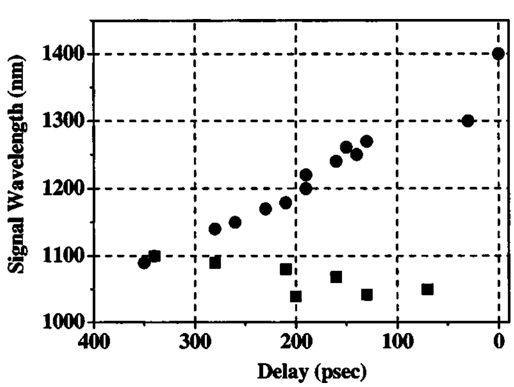

FIG. 3. Delay between the output from the Si photodiode and the signal from the InGaAs photodiode as a function of the wavelength. Lower signal's lobe—squares. Upper signal's lobe—circles.

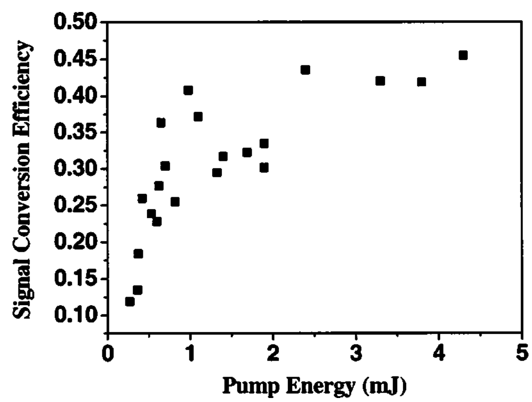

FIG. 5. Signal conversion efficiency vs pump energy (compensated for Fresnel and filter losses).

poled crystals is demonstrated. When pumped by a chirped Ti:sapphire laser with 12 nm FWHM bandwidth, a 350 nm bandwidth originating at 1100 nm was obtained in PPKTP ($\Delta\omega/\omega = 0.27$, corresponding to $\Delta\omega/\omega = 0.45$ at the idler). By choosing an appropriately poled crystal and phase-matching configuration, the IR spectrum up to 5.5 μm can be covered with ultrawide-band chirp. Future development in poling other crystals may extend this ability to the far infrared.

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