**TECHNICAL NOTE**

# Broadly tunable femtosecond pulse generation in the near and mid-infrared

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High-repetition-rate (80-MHz) femtosecond infrared pulses are generated by difference frequency mixing (DFM) a femtosecond Ti:sapphire laser with a phase-locked synchronized cw mode-locked Nd:YAG picosecond laser. This DFM scheme is of particular interest for generating ultrashort near-IR pulses (~10 fs) because group velocity mismatch with a pump pulse can be ignored. The simplicity and the broad wavelength tunability (from the near IR to the mid-IR) of this scheme is demonstrated. Short (125-fs FWHM) optical pulses in the near IR around 1.5 μm are obtained with noncritical type-I phase-matched $LiB_3O_5$. We also used a similar scheme to generate mid-infrared pulses at 3.0 μm with type-II phase-matched $KTiOPO_4$. © 1997 Optical Society of America

The considerable activity in femtosecond infrared laser development is motivated by the utility of such sources in fundamental and applied studies in optoelectronics and optical fiber communications (1–2 μm), material sciences and time-resolved chemistry and biology (3–12 μm). An important goal is to develop simple, yet broadly tunable, high-repetition-rate femtosecond infrared (IR) sources. This goal has been achieved largely with the development of femtosecond optical parametric oscillators (OPO's) based on colliding pulse mode-locked[1,2] and Ti:sapphire (Ti:Sa) laser oscillators.[3,4] OPO's in the 3–5-μm range were also demonstrated recently.[5,6] An alternative scheme for high-repetition-rate femtosecond IR generation is based on difference frequency mixing (DFM). These schemes are usually optimized to cover either the near IR or mid-IR. In the near IR, DFM was originally developed from synchronously pumped Nd:YAG systems.[7] In the mid-IR, progress has been made with Ti:Sa oscillators. One approach implemented DFM of two colors from a dual-cavity Ti:Sa laser.[8] Other methods employed DFM of signal and idler outputs from a Ti:Sa pumped OPO.[9,10] With the development of 10-fs Ti:Sa lasers, it is now possible to develop methods to downconvert such pulses to the near IR while maintaining this very short pulse duration. This can be achieved, in principle, using DFM with a phase-locked picosecond pulse in thin noncritically phase-matched crystals, as discussed below. We estimate that such pulses should be slightly shorter than those expected from currently available OPO's because group velocity mismatch with a pump beam (picosecond duration) can be ignored.

Here we describe a simple, broadly tunable, high-repetition-rate femtosecond IR source based on DFM that covers both the near-IR (with temperature-tuned LBO) and mid-IR ranges (with angle-tuned KTP). This source is derived from phase-locked synchronization of a femtosecond Ti:Sa laser with a mode-locked picosecond Nd:YAG laser. The generation of high-power, low-repetition-rate 100-fs near-IR pulses, based on amplification of synchronized oscillators, was demonstrated recently.[11] A similar scheme that produces high-repetition-rate, somewhat longer (200-fs) IR pulses was also shown recently.[12] The synchronized Nd:YAG laser allows for simple, external DFM generation of mid-IR pulses using the 1.064-μm fundamental and near-IR pulses using the 532-nm second harmonic. The tuning of the Ti:Sa laser together with angle or temperature tuning of the DFM crystal yields broadly tunable IR pulses. Since no cavity (with requisite intracavity dispersion control) is required, changing from the near IR to the mid-IR simply involves replacing the DFM crystal.

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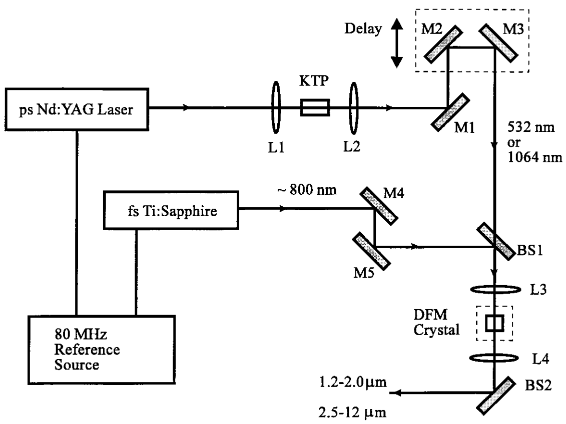


Fig. 1. Experimental setup for the generation of broadly tunable femtosecond infrared pulses with a phase-locked DFM scheme: L1–L4, plano–convex lenses; M1–M4, steering mirrors; BS1, BS2, dichroic mirrors. The DFM of 1.064 μm with Ti:Sa produces, in principle, a mid-IR range of 2.5–12.4 μm. DFM with 532 nm produces, in principle, the 1.2–2.0-μm range. In our demonstration experiments, the DFM crystals are type-II angle-tuned KTP and type-I temperature-tuned LBO producing 3–3.6 and 1.5–1.73 μm, respectively.

The experimental setup is shown in Fig. 1. The synchronization system is based on two phase-locked loops. A Spectra-Physics Tsunami Ti:sapphire laser (100-fs pulse width) was phase locked to an external 80-MHz radio frequency oscillator. The cw mode-locked Spectra-Physics Nd:YAG laser (150-ps pulse width) was also locked (using Lightwave Electronics S1000) to the same oscillator such that the synchronization achieved between the two mode-locked laser systems had a relative timing jitter of 1–2 ps. The phase noise (timing jitter) for each laser with respect to the 80-MHz oscillator was measured with a HP3561A spectrum analyzer over a bandwidth from 0.2 Hz to 20 kHz. With a pump power of 6 W from an argon-ion laser, the Ti:Sa laser produced 500-mW output power near 800 nm. For the near-IR generation, the Nd:YAG laser (average power of 6.0 W) was frequency doubled in a 12-mm KTP crystal. The average power at 532 nm was 500 mW. A 1:1 telescope was used for second harmonic generation and incorporated two 50-mm lenses that were also used as beam parameter adjustment elements in the subsequent parametric frequency mixing stage. The temporal overlap of the 532-nm pump pulses and the tunable Ti:Sa laser pulses was achieved with an adjustable passive electronic delay line in the synchronization system. Fine temporal adjustments are also possible with an optical delay line in the pump beam. A 50-mm lens was used to focus both mixing beams after a dichroic mirror combiner, giving the tight focusing required for the frequency mixing setup. We estimated that the focal spot sizes of the 532- and 800-nm beams were 24 and 30 μm, respectively. A 1.5-mm, $x$-cut $LiB_3O_5$ (LBO) crystal was used as the nonlinear medium for the near-IR frequency mixing. Type-I noncritical phase matching for DFM was realized by temperature tuning. Because of the long pulse width at 532 nm, the group velocity mismatch (GVM) between the pump and parametric pulses is unimportant; therefore, the GVM between the femtosecond signal and the femtosecond idler becomes the major factor of concern. The GVM calculated from the Sellmeier equation[13] is ~16 fs/mm between the signal and idler fields, which indicates that a much longer crystal can be used for even higher frequency conversion efficiency. Equivalently, much shorter Ti:Sa pulses could be downconverted, as discussed below. The generated near-IR power was filtered through a dichroic mirror filter and cross correlated with the Ti:Sa laser. The pulse width of the near-IR pulses was deduced from a deconvolution of the cross-correlation measurement, as discussed below. For the mid-IR generation, the LBO crystal was simply replaced with a $KTiOPO_4$ (KTP) crystal and the Nd:YAG fundamental was used rather than the second harmonic.

For 800- and 532-nm DFM, the phase-matching temperature was measured to be 135 °C. At ~400-mW power level from both pump and signal beams, 15 μW of infrared power around 1.588 μm was obtained. Since we were not operating in the pump depletion region, the generated idler power was linearly proportional to the input pump and signal power. We estimated 2.7 mm as the confocal parameter of the 532-nm beam. In our case, the crystal length was 1.5 mm; the optimal crystal length would be closer to 3 mm. Because the crystal length is shorter than the confocal parameter of the focusing beams, we used the plane wave approximation for parametric mixing. The measured DFM infrared power (15 μW) is close to the theoretical value (17 μW) based on the plane wave approximation. The large temperature tuning bandwidth (25 °C FWHM), as shown in Fig. 2(a), greatly relaxes the crystal temperature requirements and also eliminates the need for continuous temperature adjustments when the signal frequency is changed. The broad phase-matching bandwidth (50 nm FWHM), shown in Fig. 2(b), demonstrates the continuous wavelength tuning capabilities of the current scheme while keeping the crystal temperature constant at 135 °C. The cross correlation (CC) of the generated near-IR pulses with the 100-fs Ti:Sa pulses is shown in Fig. 3(a). The crystal used for the CC measurements was 0.4-mm KDP and, hence, GVM in the CC crystal was negligible. The CC curve has a width of 176 fs FWHM, which corresponds to a temporal pulse width of 125 fs FWHM for the DFM 1.5-μm pulse. (A sech-squared pulse shape has been assumed for both the Ti:Sa and DFM pulses.) The GVM, calculated from the Sellmeier equation, between the generated near-IR and the Ti:Sa pulses for a 1.5-mm LBO is ~24 fs, which is consistent with the measured temporal broadening from 100 to 125 fs. It is interesting to note that group velocity dispersions (GVD's) are negligible (~0.14 fs/nm mm at 800 nm and ~0.05 fs/nm mm at 1.58 μm).

The measured optical spectrum of the generated

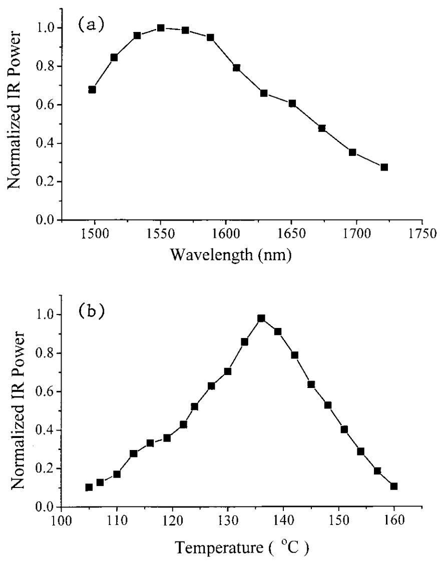


Fig. 2. (a) Wavelength tuning curve of LBO crystal for generation of 1.588-μm femtosecond pulses at $T = 135$ °C. (b) Temperature tuning curve of LBO crystal for generation of 1.588-μm femtosecond pulses.

1.588-μm near-IR pulse is shown in Fig. 3(b). The measured spectral bandwidth of the DFM pulse was 42 nm FWHM. This agrees reasonably well with that expected based on the 12-nm FWHM bandwidth of the 800-nm pulse. This means nearly all the spectral components of the signal pulse are involved in the parametric frequency mixing process. We calculate the time–bandwidth product to be 0.62, suggesting some frequency chirp. Most of this chirp was introduced in the downconversion process because of GVM in the 1.5-mm long crystal.

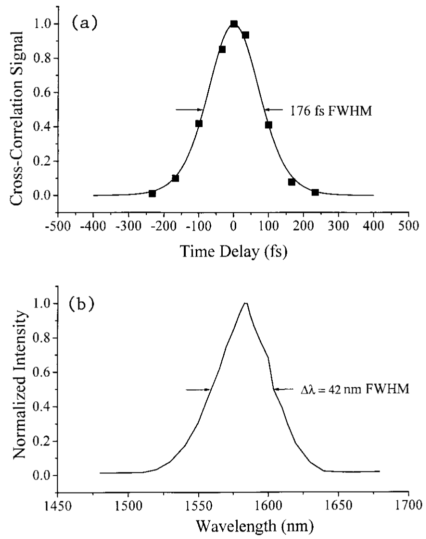


Fig. 3. (a) Cross correlation between the output infrared pulses at 1.5 μm and the 100-fs pulses from the Ti:Sa laser at 800 nm. The solid curve is a fit using sech-squared function pulses for the 1.5-μm and 800-nm beams. The deconvoluted 1.5-μm pulse duration is 125 fs. (b) Spectrum of the output DFM signal from LBO crystal in the near infrared (1.5 μm) obtained with an optical spectrum analyzer (HP 70950A). The observed near-IR spectrum agrees well with that expected from downconversion of the idler (Ti:Sa) pulse spectrum.

The frequency range of our experimental arrangement (Fig. 1) was further extended to the mid-IR by DFM of the Ti:Sa with the 1.064-μm laser beams. This scheme can, in principle, generate tunable femtosecond pulses between 2.5 and 12.4 μm. In preliminary measurements, we generated 3.0-μm femtosecond pulses, tunable in the 3.0–3.6-μm range, simply by tuning the Ti:Sa laser over the 750–820-nm range. By changing the optics set in the Ti:Sa laser, longer wavelength ranges can be obtained. We used a KTP crystal (type-II, 40.4° cut) as the nonlinear medium in place of the LBO crystal. With an input power of 3 W (1.064 μm) from the Nd:YAG we were able to obtain 10 μW at 3.0 μm. The current limitations, resulting from transmission cutoff of KTP (4.5 μm), can be addressed by using other DFM crystals (e.g., proustite).

In conclusion, we have described a simple, broadly tunable, femtosecond source of near-IR and mid-IR radiation based on external DFM of synchronized oscillators. This source is stable, easy to tune, and should cover both the near-IR (1.2–2.1 μm) and the mid-IR (2.5–12.4-μm) ranges simply by replacing the external DFM crystals. One interesting aspect of the near-IR DFM scheme presented here (using noncritically phase-matched LBO) is that the GVM and the GVD are small and the temperature phase-matching bandwidth is broad. Furthermore, because of the long (picosecond) duration of the synchronized pump pulse, the GVM with the pump pulse can be ignored. This suggests that, with thin crystals (e.g., $<0.3$ mm), the entire spectrum of a 10-fs Ti:Sa pulse can be downconverted to the near IR with minimal pulse broadening. Such a pulse would have only a couple of optical cycles and would cover most of the spectral region between 1 and 2 μm. We estimate that infrared pulses generated in this way should be slighly shorter than those derived from 800-nm pumped type-II BBO optical parametric oscillators: the GVM between the 800-nm signal and the 1.5-μm idler in type-I LBO is 16 fs/mm, compared with the GVM between the 800-nm pump and 1.5-μm signal in type II BBO of 20 fs/mm. Furthermore, a synchronously pumped OPO cavity operating around 1.5 μm would require extensive consideration of higher-order dispersion control in order to support 10-fs pulses. We suggest that, because of its inherent simplicity, DFM with a synchronized picosecond laser is a practical low-power alternative.

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