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# High power tunable femtosecond visible and infrared light from a synchronized Ti:sapphire/Nd:YAG laser system by difference frequency mixing

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**Abstract**

High power, 20 Hz repetition rate difference frequency mixing of externally synchronized femtosecond Ti:sapphire laser radiation with harmonics of a phase locked picosecond Nd:YAG laser, in a BBO crystal, is shown to produce femtosecond pulses in the visible (666 nm) and infrared (1.77 $\mu$m) regions. Using 50 $\mu$J of 760 nm amplified Ti:sapphire light, outputs of tens of microjoules at 666 nm, or several microjoules at 1.77 $\mu$m, were achieved. This technique can be used to extend the tuning range (720–1000 nm) of the Ti:sapphire laser into the visible (550–700 nm) and the infrared (1.1–2.0 $\mu$m) regions of the spectrum.

## 1. Introduction

The commercial availability of titanium sapphire lasers has introduced a broadly tunable femtosecond laser source into many laboratories. However, the tuning range (720–1000 nm) is still too restrictive for many applications. Even with harmonic generation, there are "windows" in the visible spectrum, and infrared wavelengths are not possible, especially in the important 1.3–1.5 $\mu$m range where zero dispersion optical fibers and related optoelectronic devices operate. Extending the tuning range of femtosecond Ti:sapphire lasers will be important in many areas of physics, biology, materials science and chemistry. Our particular interest in broadly tunable femtosecond pulses is in the study of gas phase chemical reaction dynamics where low repetition rate, microjoule-energy, tunable pulses in the visible are required.

Recent developments in phase locking technologies permit the synchronization of both actively and passively mode-locked lasers (see, for example, Ref. [1]) with external reference oscillators. In particular, we consider the phase locking of a femtosecond Ti:sapphire laser with a picosecond Nd:YAG laser; the synchronization of the two optical pulse trains allows us to investigate new Ti:sapphire wavelength conversion schemes based upon sum or difference three-wave frequency mixing of Ti:sapphire fundamental (or second harmonic) radiation with Nd:YAG fundamental and harmonics. Table 1 shows the ranges of wavelengths which are possible in principle using a technique which takes advantage of the synchronization of a Ti:sapphire laser with a Nd:YAG laser. We demonstrate only two of these schemes; clearly, each scheme will have very different requirements for crystals, etc.

Several techniques are available to produce femtosecond infrared radiation from Ti:sapphire lasers,


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Table 1
Table of wavelengths which are possible from sum and difference frequency mixing Ti:sapphire pulses with harmonics of Nd:YAG pulses

| Scheme | Min $\lambda$ | Max $\lambda$ |
|---|---|---|
| $\omega + 266$ | 195 | 210 |
| $2\omega + 532$ | 215 | 258 |
| $\omega + 355$ | 238 | 262 |
| $2\omega + 1064$ | 270 | 340 |
| $\omega + 532$ | 305 | 345 |
| $2\omega$ | 360 | 500 |
| $266 - \omega$ | 362 | 420 |
| $\omega + 1064$ | 430 | 515 |
| $2\omega - 1064$ | 545 | 940 |
| $355 - \omega$ | 550 | 700 |
| $\omega$ | 720 | 1000 |
| $2\omega - 532$ | 1115 | 8300 |
| $532 - \omega$ | 1140 | 2035 |
| $355 - 2\omega$ | 1225 | 25500 |
| $\omega - 1064$ | 2230 | 16600 |

$\omega$ is the Ti:sapphire fundamental. Wavelengths are in nm.

optical parametric oscillators (OPO's) being the best known [2]. Travelling wave OPO's (TOPO's) can be used with an amplified Ti:sapphire output to shift the wavelength to the infrared [3,4]. Difference frequency mixing of CPM lasers with frequency-doubled Nd:YAG light has been demonstrated [5] although the output is not broadly tunable. Subpicosecond infrared pulses between 1.4 and 1.6 $\mu$m have been generated by mixing the output of a cavity dumped synchronously pumped dye laser with the 1.064 $\mu$m output of the Nd:YAG pump laser [6]. However, in these cases low energy pulses at high repetition rates ($\sim$ 80 MHz) were produced, that are not suited for applications in gas-phase chemistry. The production of high energy pulses at low repetition rate (10–20 Hz) in the infrared by difference frequency mixing was demonstrated only for nanosecond pulses [7,8]. In this paper we show that difference frequency mixing can be used with synchronized femtosecond Ti:sapphire and picosecond Nd:YAG lasers to provide tunable visible and infrared femtosecond radiation at $\mu$J energy levels, well suited for experiments in chemistry.

## 2. Experimental arrangement

The experimental arrangement is shown in Fig. 1. A Spectra-Physics "Lok-to-Clok" Tsunami Ti:sapphire laser pumped with a 6 W argon-ion laser was used to generate 150 fs pulses at 80 MHz repetition rate at a wavelength of 760 nm. For our high power application, the Ti:sapphire output was amplified by a three-stage prism dye cell amplifier chain, using LDS-751 dye pumped with 10 mJ of synchronized picosecond 532 nm light (a Ti:sapphire regenerative amplifier could also have been used). Two further three-stage dye amplifier chains, not used in the present experiment, can be used to amplify different wavelengths for pump-probe experiments. The amplified 760 nm light had a pulse energy of up to 500 $\mu$J, and upon recompression, had a pulse duration of $\sim$ 150 fs. In the present experiment, the amplified spontaneous emission (ASE) energy was less than 1%. The "Lok-to-Clok" electronics of the Ti:sapphire laser synchronized the optical pulse train to an external 80 MHz reference oscillator.

A Lightwave Electronics Model 131 mode-locked diode-pumped picosecond Nd:YAG laser was likewise synchronized to the same 80 MHz reference source through its internal phase-locking electronics. The two optical pulse trains were thus synchronized to within a few picoseconds, as measured by optical cross-correlation. The diode-pumped Nd:YAG laser was modified to produce 150 ps pulses by installing an etalon in the cavity; this allowed for gain-limited rather than damage-limited amplification of these pulses in the following amplifier. The Nd:YAG pulses were amplified in a Positive Light RGN Nd:YAG regenerative amplifier operating at 20 Hz which provided outputs at 1064 nm ($>$ 200 mJ per pulse), 532 nm (100 mJ) and 355 nm (40 mJ). Some of the 532 nm light was used to pump the dye amplifier chain.

The residual picosecond pump light (355 or 532 nm) was collimated to a 3 mm beam diameter and combined with femtosecond light of the same beam diameter. The two collinear, copropagating pulses were passed through a 5 mm thick BBO crystal cut at 26 degrees, and the relative delay was adjusted so that the two pulses temporally overlapped.

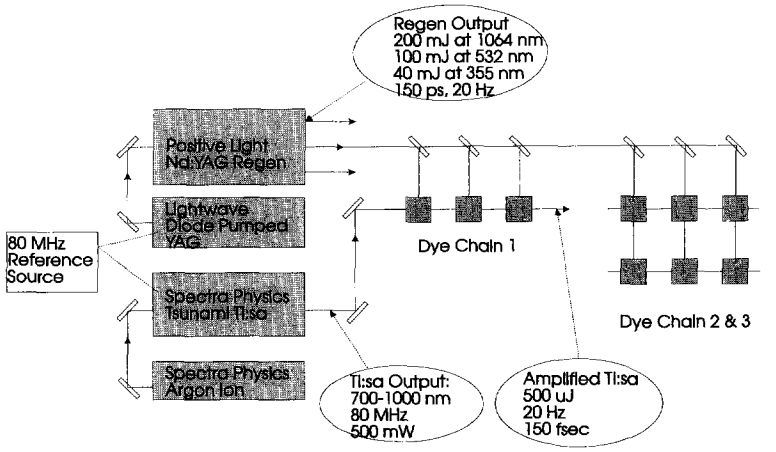


Fig. 1. Schematic diagram of the laser system. Both the femtosecond Ti:sapphire laser and the picosecond Nd:YAG laser were phase-locked to an external 80 MHz reference source. The Ti:sapphire light was amplified by a three stage dye amplifier, and the output was difference-frequency mixed in BBO with harmonics of the Nd:YAG laser.

## 3. Generation of visible light

In order to generate tunable visible light, 355 nm and 760 nm light were difference frequency mixed to give a 666 nm output. The BBO crystal optical axis was aligned vertically, the 760 nm light (signal) was polarized horizontally and the 355 nm light (pump) was polarized vertically. The resulting 666 nm (idler) was horizontally polarized for Type I phase matching at 33 degrees.

Energies were measured with a calibrated Newport 818-09J pyroelectric detector. Pump power was measured with a Molectron power meter. The 666 nm output was separated from the other wavelengths with a grating; grating reflectivity was taken into account in the energies shown below.

Group velocity dispersion (GVD) in nonlinear crystals usually limits crystal thickness to less than a millimeter for ultrashort pulses. However, with difference frequency mixing, the difference in group velocities of the signal and idler waves is quite small as compared with the pump pulse. In our case, the pump pulse duration (150 ps) was considerably longer than either the 760 nm or the 666 nm pulses; therefore the two femtosecond pulses "walked through" the picosecond pump pulse but stayed close to each other. In the presence of pump depletion, GVD actually enhances the conversion efficiency by allowing the signal and idler waves to interact with more pump photons than would be possible with zero GVD.

Table 2
Table of calculated relative group velocity time delays for the three waves used in the two difference frequency mixing schemes, in BBO

| 355 nm pump | | 532 nm pump | |
|---|---|---|---|
| 760 versus 355 nm | −273 | 760 versus 532 nm | −42 |
| 666 versus 355 nm | −243 | 1770 versus 532 nm | −85 |
| 760 versus 666 nm | 30 | 760 versus 1770 nm | 43 |

Times are in fs per mm of crystal, relative to the exit time of the pump pulse. Note that for both schemes, the GVD delay between the signal and the idler (right hand column) is quite small.

Table 2 shows the calculated delay of the two short pulses relative to the pump pulse for BBO based upon the Sellmeier equation for BBO [9]. For 5 mm of BBO, the 760 and 666 nm pulses exit 150 fs apart, so for this thick crystal there will be significant GVD broadening of the 666 nm pulse. If the 355 nm pulse were to be 150 fs in duration also, the crystal length would be limited to 500 $\mu$m.

The average intensity of the pump and signal pulses entering the crystal were $6 \times 10^8$ W/cm$^2$ and $3 \times 10^9$ W/cm$^2$, respectively. Fig. 2 shows the 666 nm idler output energy versus the 355 nm pump input energy. The input 760 nm pulse had a low-intensity component due to dye amplifier ASE, containing approximately 1% of the pulse energy and having a 150 ps duration. It can be seen that this ASE is being amplified exponentially while the femtosecond component grows linearly. This is because the much higher power

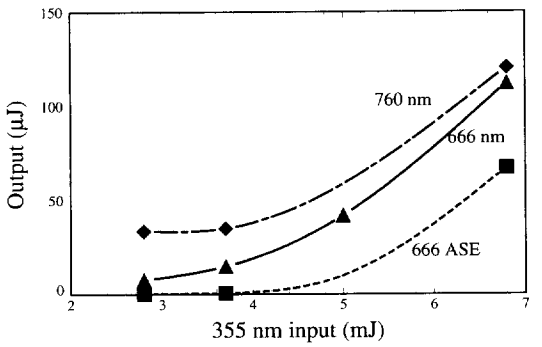


Fig. 2. Energy of difference frequency outputs versus pump pulse energy. Both the 760 nm input pulse and the generated 666 nm output pulse are amplified. Note the preferential amplification of the ASE at high pump energy.

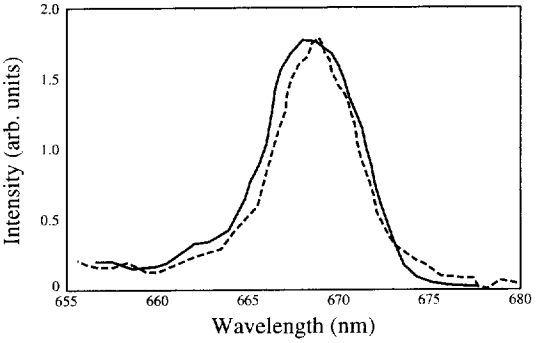


Fig. 3. Spectrum of the 666 nm difference frequency (solid line) when using the 150 fs 760 nm input. The bandwidth of the down-converted light which is expected from the measured spectrum of the input 760 nm light is also shown (dashed line); this is calculated as $S_{666}(\lambda) = S_{760}((1/355 - 1/\lambda)^{-1})$, where $S_{760}(\lambda)$ is the measured spectrum and $S_{666}$ is what is plotted as the dashed line.

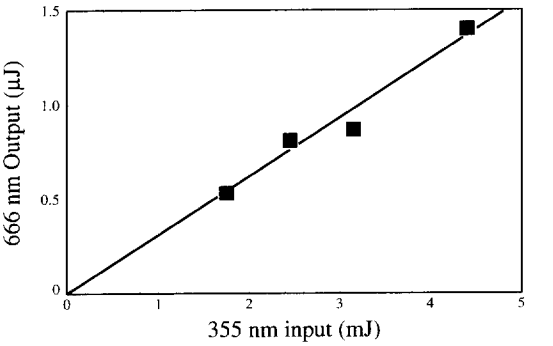


Fig. 4. Difference frequency output using a 1 mm thick BBO crystal versus pump input. The conversion efficiency is lower than with the thicker crystal.

femtosecond component saturates the conversion process by pump depletion. The ASE could be eliminated by introducing a saturable absorber into the dye amplifier chain. We suggest that it would be advantageous to stretch the 760 nm signal pulse upon entering the crystal, and recompress the idler pulse afterwards. This would enhance the idler generation by optimally overlapping in time the signal and pump pulses.

Also presented in Fig. 2 is the 760 nm pulse energy after the crystal, showing that this light is being amplified, along with the 666 nm light. A parametric gain in the 760 nm light is expected. At a pump energy of 4 mJ, the conversion efficiency of the pump light during the 1.25 ps interaction in the crystal is measured to be $\sim 40\%$.

The pulse duration of the 666 nm light was measured with a single-shot autocorrelator to be 295 fsec. Since half of this value is due to GVD between the signal and the idler in the 5 mm crystal, it is not possible to further reduce the duration by compression.

The pulse spectrum was measured in a 0.2 m monochromator using a linear diode array detector to capture the entire bandwidth. The spectral width of the 666 nm pulse is shown in Fig. 3. The measured width of 6 nm agrees well with that expected from the downconversion of the measured width of the 760 nm spectrum, which is included in the figure for illustration. The FWHM bandwidth of the difference frequency phase matching in the 5 mm crystal was measured to be 15 nm, sufficient to support sub-100 fs pulses.

The conversion efficiency was also measured in a 1 mm thick BBO crystal (CSK Optronics), as shown in Fig. 4. The pump and idler intensities were the same as for the 5 mm thick crystal. The conversion efficiency ($\sim 2\%$) is lower than for the thick crystal, as expected. The pulse duration was not measured because of the lower energy, but from Table 2 we expect the effect of GVD to be minimal. In this experiment, the conversion efficiency was limited by damage of the BBO crystal due to the pump laser. This particular 1 mm crystal was not contained in ahousing which usually protects hygroscopic crystals from surface damage. We expect, therefore, that a properly housed BBO crystal should allow for improved conversion efficiency.

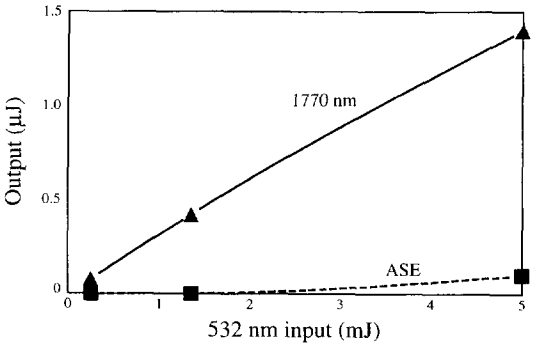


Fig. 5. Difference frequency generation of 1770 nm light versus pump energy, using a 5 mm thick BBO crystal. Also shown is the level of downconverted ASE.

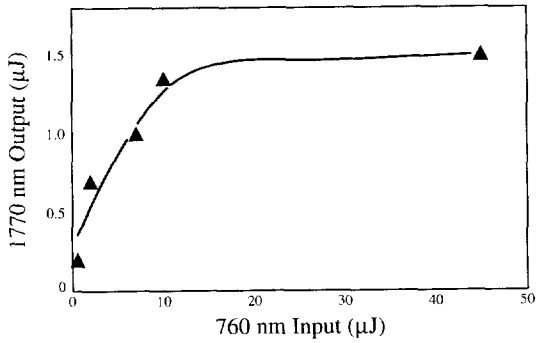


Fig. 6. Difference frequency generation of 1770 nm light versus 760 nm input energy. The saturation indicates that the process is limited by the number of pump photons available. Pre-stretching the 760 nm input should circumvent this problem and therefore improve the conversion efficiency.

## 4. Generation of infrared light

An analogous mixing scheme was used to generate femtosecond infrared pulses. In this case, some picosecond 532 nm light was used as the pump and mixed with the femtosecond 760 nm light (signal) to produce 1770 nm light (idler). A 5 mm thick BBO crystal was used with Type I phase matching, and the results are shown in Fig. 5. The 1770 nm light was separated from the shorter wavelengths by filtering with RG850 glass, and the wavelength was verified by measuring its transmission through GaAs and germanium. The calculated GVD is shown in Table 2. The sensitivity of the 1770 nm energy to the 760 nm input is shown in Fig. 6, and it again appears that the process is limited by the number of pump photons.

This same process might be used to generate 1.5 $\mu$m light by using a 824 nm Ti:sapphire wavelength, or 1.3 $\mu$m by using 900 nm. Longer wavelengths might be obtained by difference frequency mixing the Ti:sapphire light with the 1064 nm output from the Nd:YAG laser. Difference frequency mixing of two unsynchronized 82 MHz femtosecond Ti:sapphire lasers in KTP has recently been demonstrated [10]. We intend to investigate the mixing, at 80 MHz, of the unamplified Ti:sapphire pulses with synchronized picosecond 532 nm pulses to generate broadly tunable 1.5 $\mu$m pulses at high repetition rate. Such a laser source could be useful for experiments in optoelectronics.

In conclusion, we have demonstrated that high power, low repetition rate difference frequency mixing with synchronized picosecond Nd:YAG harmonics can be used to shift the output of a femtosecond Ti:sapphire laser into the visible and infrared regions of the spectrum with good efficiency. The bandwidth of the difference frequency mixing technique is much greater than that of second harmonic generation. As the pump pulse duration is much longer than the femtosecond pulses, and the dispersion between signal and idler is minimal, GVD broadening and walkoff are reduced and, hence, longer crystal lengths are possible. Even higher efficiencies and OPA gain should be possible by optimally stretching the Ti:sapphire pulse before the mixing crystal.

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