Visible 20-femtosecond pulse
generation by double-pass
non-collinear optical parametric
amplifier

S. K. Karthick Kumar¹, T. Goswami¹,
I. Bhattacharyya¹ and D. Goswami²,*

¹Department of Chemistry, and
²Center for Laser Technology, Indian Institute of Technology,
Kanpur 208 016, India

We report a new design and construction of a high-
power, white light seeded double-pass non-collinear
optical parametric amplifier (NOPA) in the visible
wavelength using a single amplifier crystal. For its
successful implementation, many important practical
considerations are discussed in detail. Tunable femto-
second pulses were obtained with pulse widths less
than 20 fs and characterized using frequency-resolved
optical gating technique. The generated visible pulse,
tunable from 490 to 740 nm from the NOPA, was fur-
ther optimized by second harmonic and four-wave
mixing signals.

Keywords: Light pulse, optical parametric amplifier,
parametric gain, ultrafast spectroscopy.

OUR capability to observe fast dynamical processes is
limited by the duration of the light pulses available to us,
thus calling for the generation of shorter and shorter
pulses. However, the need to resonantly excite a system
and probe the optical transitions taking place at different
photon energies requires wavelength tunability of both
pump pulse (the one exciting the system) and probe pulse
(the one measuring the result). Therefore, both wave-
lengh tunable as well as shorter duration pulse are esen-
tial for a variety of experiments in ultrafast spectroscopy.
Since many ultrafast spectroscopy experiments do not
make use of all the output power from the multiple-stage
ultrafast optical parametric amplifiers (OPAs), it is desir-
able to build comparatively low-power non-collinear
optical parametric amplifiers (NOPAs) using off-the-shelf
components available in typical ultrafast laser laborato-
ries at a relatively lower cost. Sub 20 fs NOPA techno-
lology has become one of the active areas of research in
femtosecond lasers due to its short pulse duration, better
stability, ease of construction and easy day-to-day align-
ment over the OPAs, since the typical OPAs have mul-
tiple stages of amplification, making quick realignment
a non-trivial task. The design of the NOPA presented here
uses two passes of the pump pulse in the single amplifying
β-barium borate (BBO) crystal, with the signal gener-
ated in the first pass being used as a seed pulse in the

*For correspondence. (e-mail: dgoswami@iitk.ac.in)
to 666 nm, pumped by the second harmonic of the 2 watt Ti:sapphire oscillator and using a six-mirror folded ring cavity\textsuperscript{1}. Pang et al.\textsuperscript{3} discussed the working of NOPA theoretically. Wilhelm et al.\textsuperscript{2} used the idea to build the first sub 20 fs NOPA, and Cerullo et al.\textsuperscript{4} built the first sub 8 fs NOPA with 180 THz bandwidth and 2 µJ output pulse energy. Baltuska et al.\textsuperscript{2} built a 4 fs NOPA using programmable dispersion control. Conventionally, for the two-stage NOPA, only a small fraction of the blue pump was used as a pump in the first pass and the rest was used to pump the amplified signal generated in the first pass in a separate amplifier crystal to get high power tunable pulses. Tan et al.\textsuperscript{6} chirped the blue pump pulse and demonstrated that the chirp can be transferred to the amplified output pulse. Mohring et al.\textsuperscript{7} and von Vacano et al.\textsuperscript{8} shaped the white light supercontinuum seed using a liquid crystal modulator, which indirectly shaped the output pulses in the visible. Also, they generated the white light seed using a photonic crystal fibre with a pulse shaper to produce highly stable and shaped tunable optical pulses. Wasylyczyn et al.\textsuperscript{9} built a NOPA with a single BBO crystal design, which generated the second harmonic as well as the resulting amplification.

One can understand the main principle behind the group velocity dispersion by expanding the wave-vector mismatch $\Delta k$ in the output powers of the seed wavelength detuning $\Delta \lambda$ to get\textsuperscript{1}:

$$\Delta k = \Delta k_0 + \frac{\partial \Delta k}{\partial \lambda_1} \Delta \lambda_1 + \frac{\partial^2 \Delta k}{\partial \lambda_1^2} \Delta \lambda_1^2 + \cdots.$$ 

For a vanishing value of $\partial \Delta k/\partial \lambda_1$ and to satisfy the phase matching condition $\Delta k_0 = 0$, it requires that,

$$v_{g,1} = \cos(\Omega) v_{g,2},$$

where $v_{g,1}$ and $v_{g,2}$ are the group velocities of the signal seeded and the generated idler pulses respectively, and $\Omega$ is the angle between them\textsuperscript{1}. In the process of parametric amplification of the signal by the amplifying pulse, the idler pulse is generated, which is at the difference frequency between the signal and amplifying pulse. A finite group velocity mismatch between the signal and the idler pulses arises as the idler pulse walks faster along the amplifying pump pulse, while the seed signal pulse lags behind. Because of this, a lengthening of the pulse occurs. However, in the non-collinear geometry, only the projection of the idler group velocity is on the seed, which is equal to the signal group velocity as discussed in the above case, and as such no lengthening of the pulse occurs. Thus, long BBO crystals can also be used to generate shorter pulses for NOPAs with high efficiency, which is in contrast to the OPAs that require thinner and thinner BBOs for obtaining relatively shorter pulses with lower and lower efficiency.

To begin with, it is desirable to build a NOPA on an optical breadboard to gain extra stability with careful choice of mounts, holders and stages. Second harmonic of the fundamental from the ultrafast Ti:sapphire amplifiers like regenerative, multipass or cavity-dumped amplifier can be used to generate the pump for the NOPA. When using regenerative and multipass ultrafast amplifiers, input pulse energy of 100–200 µJ at 1 kHz is required. In our set-up as shown in Figure 1, we use 200 µJ of vertically polarized fundamental beam from Ti:sapphire amplifier with central wavelength of 800 nm. We split this fundamental beam with a thin 90 : 10 UVFS beamsplitter (CVI, BS-1-800-10-1012-45-P) and the transmitted 180 µJ part was used to produce the second harmonic blue pump pulse. To avoid any damage to the second harmonic generating (SHG) BBO crystal, the fundamental beam was not focused onto the BBO crystal to produce the blue pump. Instead, the beam diameter was reduced to ~1.5 mm using a Galilean telescope arrangement consisting of a convex lens of 200 mm focal length and a concave lens of ~150 mm focal length, and then sent into the SHG crystal. Depending on the input beam diameter and the power available from the Ti:sapphire amplifier for the NOPA, other ratios of focal lengths of the lenses in the telescope would be more suitable. If the available power from the ultrafast amplifier is around 100 µJ, then the fundamental 800 nm pulse can also be focused just behind the SHG crystal and the output diverging blue beam can be collimated using an anti-reflection (AR) coated lens. A type-1 BBO crystal AR-coated for 800 nm, 1 mm thick, cut at $\theta = 29^\circ$ was used for SHG, as it was found to produce a good quality spatial mode in the beam profile of the output beam rather than using a thicker SHG BBO for higher conversion efficiency. It was observed that the good beam quality (checked at a far field) of the generated SHG is far important than its power. In fact, in our case, the 1 mm SHG BBO crystal was found to provide a good quality beam compared to a 2 mm thick BBO crystal. A poor mode of the SHG beam reduces the parametric gain in the second BBO crystal used for amplification. Alternatively one can use a pair of cylindrical lenses AR-coated for 400 nm, to shape the SHG beam.

We used the reflected beam from the 90 : 10 beamsplitter to generate a single filament white light continuum as our weak signal. We used a convex lens of 50 mm focal length to focus the 20 µJ part of the Ti:sapphire laser onto a 2 mm thick sapphire plate (Newport, 10SW8-180) to produce white light continuum (circular beam without any coloured rings). The spectrum of white light was flat from 460 to 700 nm, when generated in a sapphire plate. To control the white light continuum, an iris was used to reduce the intensity at the sapphire plate until a good-quality single-filament white light continuum was observed. Alternatively, a variable attenuator can also be used. It is important to place the sapphire plate on a rotation

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mount (Newport, RM25-A), which will allow us to focus the beam at different portions of the plate if the focusing spot gets damaged accidentally or by prolonged use. The sapphire plate was also mounted on a translation stage to finely vary its position around the focal point. The white light was monitored by a photodiode for pulse-to-pulse stability. This white light beam was routed around to match the path length to that of the pump. As shown in Figure 1, the generated white light was focused by a lens L2 (f = 50 mm) a few millimetres behind a 2 mm-thick, broadband AR-coated, type 1 BBO crystal cut at θ = 26° (amplifier crystal, CASTECH). The SHG blue pump was passed through a 400 nm AR-coated, 15 mm thick, UV fused-silica plate to stretch the pulse to a few hundreds of femtosecond width. The use of such a plate was necessary for this particular set-up, since our input pulses from the multipass amplifier are very short (typically, 45 fs). In case of a regenerative amplifier, for example, this plate need not be used. The blue pulses should be stretched as this will ensure more temporal overlap with the highly chirped white light continuum seed, resulting in a large bandwidth gain. The stretched blue pulse was then routed through a 400 nm dielectric mirror, placed on a translation stage, which also filters the fundamental 800 nm pulse. The blue pump was loosely focused on the same spot in the amplifier BBO crystal where the white light was also being loosely focused. The amplifier crystal was placed on rotation and translation stages. A bright superfluorescence ring (emanating as a cone) in the visible can be observed after the amplifier BBO (Figure 2a). It is important that the BBO crystal be oriented horizontally along its optic axis to generate superfluorescence since the pump pulse is horizontally polarized. If superfluorescence was not observed, the pump focus can be slightly adjusted and the BBO can be slightly rotated along the vertical axis. However, when the above method fails, the distance between the gratings of the compressor in the ultrafast amplifier can be adjusted to obtain and maximize the intensity of the superfluorescence rings. If there is an excess leakage of residual 800 nm beam along the blue pump or seed beams, one can observe multicoloured rings after the amplifier BBO crystal, which might eventually damage the focused spot on the crystal. These supercontinuum multicoloured rings are due to excess
power of the light pulse used. The coloured rings seen after the amplifier BBO crystal can be ensured as the super-
fluorescence rings, if the ring diameters change drastically with the rotation of the crystal along its vertical axis, which is due to varying phase-matching angle while rotating the crystal. In case of the coloured rings generated due to supercontinuum, they remain invariant under
crystal rotation.

The white light seed and the blue pump are crossed at
an angle of 6.4°, as can be seen in Figure 1. At this angle, the white light can be seen along the circumference of the superfluorescence ring and this is a good guiding factor to ensure correct alignment. If the white light is not seen on the circumference of the superfluorescence ring, the amplifier BBO can be rotated slightly along its vertical axis to change the superfluorescence ring diameter in order to overlap with the white light seed. The delay between the seed and the blue pump can be set to zero by adjusting the translation stage, referred to as wavelength tuning stage in Figure 1. While adjusting the delay with the translation stage, one can obtain a bright signal as shown in Figure 2c and d. The idler (in the infrared region) can be easily seen on a screen in front of the ampli-
crystal around the zero delay, through an infrared viewer or using a webcam (with IR filter removed) while adjusting the delay. The idler beam appears on the left side of the ring and the signal, which is the amplification of the seed, on the right side of the ring, along the seed. While moving the translation stage, once the idler is located, one can fix the translation stage at that position. We only need to move it slightly around that delay posi-
tion to locate the signal which might appear weak at first. At this point the spatial overlap of the white light seed and the pump can be checked at the crystal. The focal volume of the seed pulse has to be contained within the focal volume of the pump, which can be easily checked using a pinhole. Also, it should be ensured that the seed and the pump remain spatially overlapped inside the cry-
atal even while the delay stage position is changed. A simple method to quickly find the zero delay would be to route the 10% beam hitting the sapphire plate, through a hollow retro-reflector on a long translation stage. Also, the pump power can be increased carefully for a brief time, while looking for the signal. Once the signal is obtained, it can be maximized by adjusting the wave-
lengtuning translation stage. Also, it is important to have the lens L2 on a XYZ lens mount (Newport, LP-1A), which will allow one to finely adjust the spatial overlap between the seed and the pump and also change the focal point of the white light. A poor overlap between the seed and pump pulse can give rise to a signal with structures as shown in Figure 2b. One can observe that the super-
fluorescence ring has completely disappeared, as shown in Figure 2d, when the green 550 nm amplified signal pulse is generated. This is because of “complete phase matching” for a large amplification bandwidth, which can give rise to very short pulses after compression. Incom-
plete phase matching can give rise to poor bandwidths as
can be seen, for example, in Figure 2e, where a 600 nm red signal pulse is seen at the right side, while the green
ring of the superfluorescence is still present. When the angle between the seed and the pump, and the crystal angle are slightly adjusted, all the colours in the super-
fluorescence disappear and the output signal pulse spe-
trum broadens (Figure 2d). Poor mode of the output signal as in Figure 2b can be avoided by adjusting the
XYZ of the lens L2 for a good spatial overlap. Fine adjust-
ment of the pump and seed focus is also necessary while trying to obtain higher power of the signal. The generated signal beam and the remaining blue pump were sent back to the amplifier BBO such that it overlaps on the BBO at a different spot, slightly below the first pass spot on the crystal, using silver concave mirrors of focal length 150 mm. The delay between them was changed by moving one of the concave mirrors on a translation stage and the generated signal in the first pass was further amplified by the second pass blue pump. The generated pulse is easily tunable from 490 to 740 nm by moving the wave-
lengtuning translation stage, as mentioned in Figure 1. The amplified output from the second pass was collima-
ted using the silver concave mirrors of focal lengths 100 (Newport, 10DC200ER.2) and 50 mm (10DC100ER.2). The NOPA output is highly chirped because the white light seed is highly chirped. So, it needs to be compressed further by a pair of prisms. We used a pair of SF10 Brew-
erster angle dispersion prisms (Newport, 10SF10) in mini-
um deviation angle, mounted on a prism stage (Newport, UGP-1) and translation stage (Newport, 423). A periscope arrangement was used to rotate the polarization of the output from NOPA as the prisms needed a particu-
ticular polarization for efficient dispersion. We found that the tip-to-tip distance between the prisms was optimal at around 200 mm for 550 nm wavelength. The distance between the prisms depends on the central wavelength of the NOPA output beam, material dispersion in the path of the seed and amplified signal, and the prism material. A folding mirror was used to send the beam back through the prism pair at a slightly lower height to pick-off the compressed output. The beam should hit the first prism at the tip while the second can be inserted more into the beam path to optimize the pulse width. A practically useful, step-by-step procedure for pulse compression using a pair of prisms can be found in Newport prism compressor application notes. To get a shorter sub 20 fs pulse, one can rotate the prism for minimum deviation and adjust the distance for every central wavelength. It will be difficult to compress the chirped output pulse from the second pass of the NOPA if the material dispersion in the path of the seed and signal is more, and this can be avoided using thin sapphire plate for white light generation and focusing the white light on the amplifier crystal using either a concave mirror or a parabolic mirror.
Typically, for a visible NOPA, its amplified compressed output is set at 600 nm through the prism compressor and is focused on to a type I SHG crystal, cut at $\theta = 45^\circ$, to generate second harmonic at 300 nm, which is quantified by a UV photodiode. The output SHG signal is then maximized by changing the distance between the prism pair and the second prism insertion before using an autocorrelation to measure the pulse width.

In our case, we maximized the second harmonic of the 500 nm beam generated in a 4 mm thick BBO crystal with $\theta = 56^\circ$ (SHG phase matching angle for 500 nm). We also performed four-wave mixing experiments in CS$_2$, where the four-wave mixing signal was maximized by optimizing the prism compressor. The NOPA output pulse width was also measured by a commercial frequency resolved optical gating (FROG) device, GRENOUILLE (Swamp Optics, Inc.), by tuning the output of the NOPA to 700 nm, as shown in Figure 3. Since this particular commercial FROG device cannot measure or is not reliable for pulses less than 20 fs and less than 700 nm centre wavelength, we measured the pulse width to be around 20 fs at 700 nm. However, it is expected to be less than 20 fs if measured by an all-reflective autocorrelator with 100 $\mu$m thick BBO crystal cut at $\theta = 45^\circ$, which can be used for a broad-phase matching wavelength range from 480 to 680 nm. To obtain even shorter pulses, UVFS prisms can be used with 1 mm separation. Also, the non-collinear angle, phase matching angle and the seed delay have to be changed for every central wavelength to maximize the signal and for short pulse generation. The amplified idler pulse (870–1500 nm) can also be compressed to obtain infrared shorter pulses. We obtained a maximum of 20 $\mu$J of pulse energy at 520 nm from the NOPA, which is presently being used for photon echo and two-dimensional electronic spectroscopy that require stable and short pulses.

We have shown that a double-pass NOPA tunable across the visible wavelengths ranging from 490 to 740 nm, can deliver transform-limited, sub-20 fs pulses with good stability (~5%) and at a smaller footprint. This is a valuable tool for ultrafast spectroscopy laboratories wanting to track dynamics at much shorter timescales. Also, constructing a NOPA can help one to understand all the other tools of the trade routinely used in any ultrafast laboratory from this single set-up.


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