PARAMETRIC NONLINEAR OPTICS

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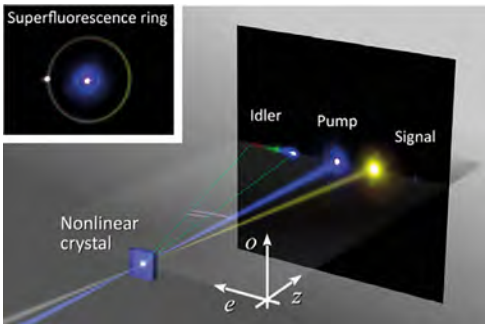

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Many scientific and technological applications require the generation of broadly tunable femtosecond light pulses. Optical parametric amplifiers (OPAs) exploit second-order nonlinear interactions to convert a high-power fixed wavelength pulse (the pump) into a tunable pulse (the signal). This paper reviews the principles of OPAs and highlights their capability to generate few-optical-cycle pulses with high energy and carrier-envelope-phase stability.

The last decades have witnessed breakthroughs in ultrafast optics, which have made sources of femtosecond light pulses broadly available to scientific and industrial users. Key enabling technologies include: i) the development of broadband solid-state optical gain media, such as Ti:sapphire and Ytterbium(Yb)-doped crystals and fibers; ii) the introduction of passive laser mode locking techniques, such as Kerr lens mode locking and semiconductor saturable absorber mirrors; iii) the invention of the chirped pulse amplification (CPA) configuration, which was awarded the Nobel Prize in Physics in 2018, and which enables to scale the energy of femtosecond pulses by many orders of magnitude.

These innovations have made available primary sources of ultrashort light pulses. Mode-locked Ti:sapphire oscillators followed by CPA with regenerative or multipass amplifiers generate short (with duration down to 20 fs) pulses at 800 nm with multi-mJ energy and 1-10 kHz repetition rate. However, their architecture is rather complex, as they require two additional green lasers to pump the oscillator and the amplifier. Yb based bulk/fiber lasers generate longer pulses (with ≈200-fs duration) but, due to the millisecond excited state lifetime of the active medium, they can be directly diode pumped and allow high repetition rates, up to several MHz. In addition, thanks to their low quantum defect, with pump wavelength at 980 nm and emission wavelength at 1030 nm, they put a low thermal load on the gain medium, allowing the generation of high average powers, up to hundreds of W. Despite their high stability and reliability, however, **primary sources only work at fixed wavelengths, with limited tunability**.

Many applications of ultrashort pulses require the ability to broadly tune the emission wavelength from the mid-infrared (MIR) to the ultraviolet (UV), to access specific light-matter interactions. This is typically achieved using the above-mentioned primary sources to drive **secondary sources, which exploit nonlinear optical effects to change the frequency, broaden the bandwidth, and shorten the duration of the output pulses.** This article provides a non-exhaustive overview of the status and perspectives of the development of tunable sources of ultrashort pulses based on nonlinear frequency conversion, focusing in particular on **optical parametric amplifiers (OPAs)**, which are powerful and versatile optical systems. The paper starts by introducing the concept of OPA, which allows for the generation of broadly tunable ultrashort pulses starting from a fixed frequency pump pulse. It then shows that OPAs, thanks to their broad gain bandwidth, can significantly shorten the pulse duration, allowing the generation of tunable few-optical-cycle pulses. It further illustrates the capabilities of OPAs, combined with the CPA architecture, to scale the pulse energies, and to control the electric-field

profile of the ultrashort pulse by stabilization of the carrier-envelope phase. The paper also discusses the possibility to further broaden the bandwidth by coherent combination of OPAs and concludes with an outlook on outstanding conceptual and technological challenges.

OPTICAL PARAMETRIC AMPLIFIERS

Nonlinear optical effects arise in matter when the amplitude of the electric field of light E is large enough that the induced polarization P becomes a nonlinear function of the field:
$P_i = \varepsilon_0[\chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \ldots]$.
where i,j,k,l are components of the electric field (polarization) vectors, $\chi_{ijk}^{(2)}$ is the second-order susceptibility tensor (which is non-zero only in non-centrosymmetric media) and $\chi_{ijkl}^{(3)}$ is the third-order susceptibility tensor (which is non-zero in any medium). Third-order nonlinear effects, also known as **four-wave-mixing processes**, are used to generate new frequencies and broaden the pulse spectrum. Particularly relevant are self-phase-modulation (SPM) in fibers and white light continuum (WLC) generation in bulk media. Second-order nonlinear effects, also known as **three-wave-mixing processes**, are used to achieve frequency conversion in non-centrosymmetric crystals. The birefringence of such crystals is often exploited to achieve the so-called **phase-matching condition**, which allows constructive interference of the nonlinear signals generated at different positions within the crystal, resulting in high frequency conversion efficiency.

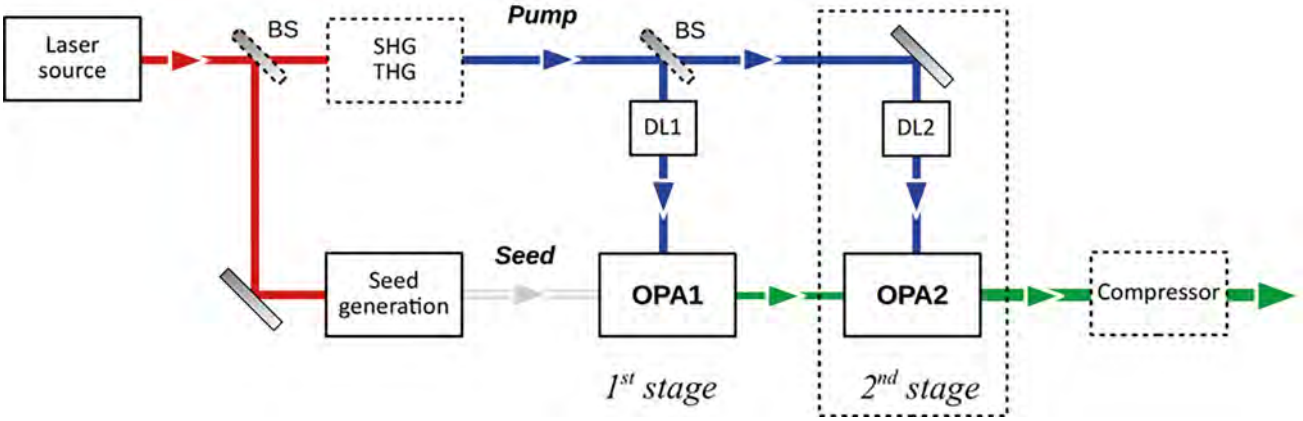
This article focuses on OPAs, whose conceptual foundations are explained in Insert 1. Briefly, an OPA is pumped by an intense pulse at frequency ω_p, the pump frequency, and amplifies a weak "seed" pulse at frequency ω_s, the signal frequency, thereby generating an idler pulse at the difference frequency $\omega_i = \omega_p - \omega_s$. An OPA therefore allows the generation of broadly tunable signal pulses starting from a powerful, fixed frequency pump pulse [1, 2]. Figure 1 illustrates the conceptual architecture of an OPA. The output of the driving laser is split into two arms: the first serves as the pump pulse, possibly following second harmonic generation (SHG) or third harmonic generation (THG); the second arm produces the seed pulse, typically by WLC generation in a sapphire or YAG plate. An optical delay line synchronizes the pump and seed pulses, since the second-order nonlinear interaction only occurs during their temporal overlap, without depositing any energy in the nonlinear medium. Parametric amplification occurs in the OPA crystal, which may be followed by one or more gain stages if needed. Finally, an optional pulse compressor corrects the spectral phase of the amplified signal pulse, achieving Fourier transform-limited (TL) pulse duration.

OPAs are typically pumped by the fundamental frequency (FF, 800 nm) or by the second harmonic (SH, 400 nm) of Ti:sapphire, or by the FF (1030 nm), the SH (515 nm) or the third harmonic (TH, 343 nm) of Yb systems. Using the most popular nonlinear crystal, β-barium borate (BBO), they achieve tunability throughout the visible and the near-infrared range (400-3000 nm). Tunability is limited in the visible by the pump wavelength and in the infrared by absorption of the nonlinear crystal. SHG and sum-frequency generation (SFG) enable extension of the tuning range to the UV, while difference-frequency generation (DFG) and optical rectification allow the generation of MIR and terahertz pulses, respectively.

FEW-OPTICAL-CYCLE PULSE GENERATION WITH OPAS

The OPA can be considered as a "photon cutter", which splits the energy of a fixed frequency pump pulse, $\hbar\omega_p$, into the sum of signal ($\hbar\omega_s$) and idler ($\hbar\omega_i$) energies. Since there are many pairs of signal and idler frequencies fulfilling the energy conservation condition, **the OPA is intrinsically a broadband amplifier, provided that the phase matching condition is satisfied for a wide range of** ● ● ●

Figure 1. Conceptual scheme of a femtosecond OPA. DL: delay line; BS: beam splitter; SHG/THG: second/third-harmonic generation module. Dashed boxes denote optional stages.



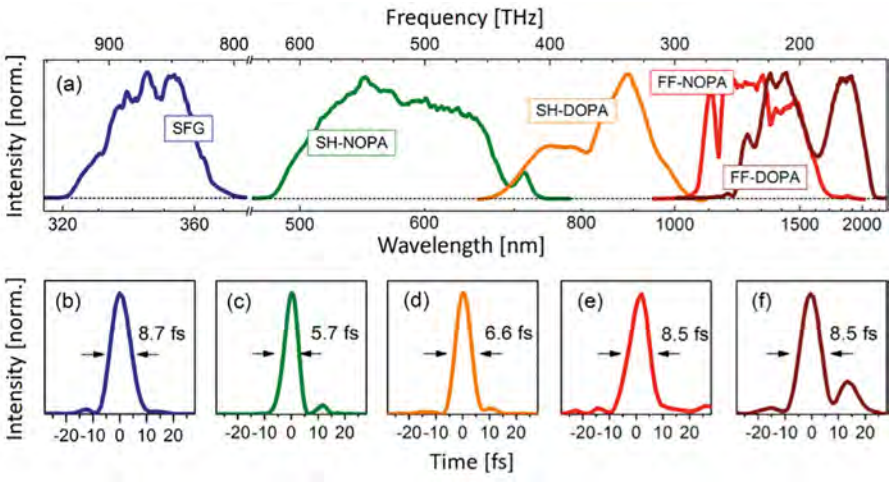


Figure 2. (a) Ultra-broadband pulse spectra generated by NOPAs and DOPAs pumped by the FF and the SH of Ti:sapphire. (b-f) Measured pulse intensity profiles; all the pulses have been compressed to sub-10-fs duration.

frequencies. To evaluate the phase matching bandwidth of an OPA, let us start from a fixed set of pump, signal and idler frequencies satisfying the phase-matching condition: $\Delta k = k_p(\bar{\omega}_p) - k_s(\bar{\omega}_s) - k_i(\bar{\omega}_i) = 0$ where Δk is the wave vector mismatch. Assuming that, for a fixed pump frequency, the signal frequency increases to $\omega_s = \bar{\omega}_s + \Delta\omega$, then the idler frequency decreases to $\omega_i = \bar{\omega}_i - \Delta\omega$ and the ensuing wave vector mismatch becomes

$$\Delta k = \left(\frac{dk}{d\omega}\right)_{\omega_s} \Delta\omega - \left(\frac{dk}{d\omega}\right)_{\omega_i} \Delta\omega = \left(\frac{1}{v_{gs}} - \frac{1}{v_{gi}}\right)\Delta\omega,$$

where v_{gs}(v_{gi}) is the group velocity of the signal(idler) pulse. This formula gives a very simple design rule: a broadband OPA requires $v_{gs} = v_{gi}$, *i.e.* group velocity matching between the signal and idler pulses [2].

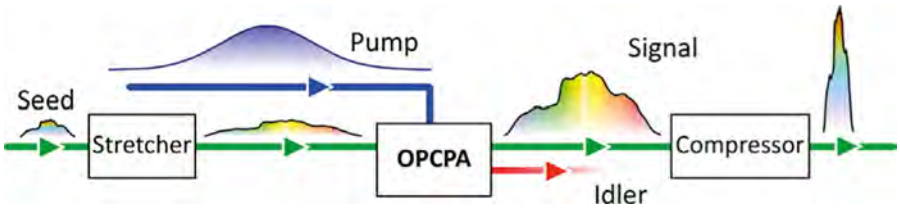
Group velocity matching is achieved when an OPA is operated around degeneracy, *i.e.* with $\omega_s = \omega_i = \omega_p/2$. In the case of a degenerate OPA (DOPA), using a type I phase matching configuration, signal and idler pulses have the same frequency and polarization, and therefore the same group velocity. Working outside of degeneracy, in general $v_{gs} \neq v_{gi}$; in this case, group velocity matching can be obtained using a non-collinear OPA (NOPA) [3]. **In the NOPA the propagation direction of the idler forms an angle Ω with the propagation direction of the signal, so that the group velocity of the signal matches the projection of the group velocity of the idler along the signal: $v_{gs} = v_{gi} \cos\Omega$.** The visible NOPA pumped by the SH of Ti:sapphire generates visible pulses with duration down to 4-5 fs and is a powerful tool for ultrafast spectroscopy of (bio)-molecules. Using NOPAs or DOPAs pumped by the FF or the SH of Ti:sapphire or by the FF, the SH or the TH of Yb, it is possible to generate, following dispersion compensation, sub-10-fs pulses broadly tunable from the visible to the near infrared, as shown in Figure 2, which reports pulses produced by NOPAs and DOPAs operated in our laboratories [4]. The figure also shows the UV pulses obtained by SFG between the visible pulse from a SH-NOPA and a fraction of the FF.

OPTICAL PARAMETRIC CHIRPED PULSE AMPLIFIERS

CPA has revolutionized ultrafast optics by allowing the scaling of the pulse energy without inducing damage in the gain medium. In a CPA system the pulse is first sent to a "stretcher", a dispersive optical system in which its frequency components acquire a relative delay, or frequency chirp, resulting in a temporal lengthening of the pulse by up to 4-5 orders of magnitude. An increase in the duration of the chirped pulse corresponds to a decrease of its intensity, so that it can now be safely amplified by several orders of magnitude without damaging the amplifier material. Following the amplification stage, the pulse is sent to a "compressor", which cancels its chirp and restores the original pulse duration. CPA systems typically use active media based on population inversion, in which energy is stored for a long time (microseconds to milliseconds) in the upper laser level. Such media can be substituted by OPAs, which offer high gain over broad bandwidths. **In the so-called optical parametric chirped pulse amplifier (OPCPA) (see Figure 3), a broadband pulse is first stretched to 1-10 ps duration, then amplified in multiple OPA crystals pumped by a long and energetic pump pulse, and finally recompressed to TL sub-10-fs duration.**

OPCPAs offer several advantages over CPA systems: i) the capability of providing high gain over a short pathlength; ii) their ultrabroad gain bandwidths in group velocity matched configurations; iii) the

Figure 3. Conceptual scheme of an OPCPA. After the stretcher, the OPCPA can consist of multiple parametric amplification stages.



With respect to active approaches, passive CEP stabilization has the advantages of the absence of electronic circuitry, of frequency tunability and of the possibility to combine ultrabroad bandwidths with energy scaling, using OPCPAs.

strongly reduced thermal load (with associated beam distortions) on the gain media, due to the lack of energy storage in the amplifying crystals; iv) the reduction of pulse pedestals due to amplified spontaneous emission. For these reasons, OPCPAs currently appear as the most promising approach to the scaling in energy and average power of few-optical-cycle pulses, up to the PW peak power level or higher [5]. There are however several challenges that need to be addressed in OPCPAs. First, suitable pump laser systems, with pulse duration from a few to a few tens of picoseconds, delivering high energies and/or high average powers, should be developed. Then, such pulses need to be electronically or all-optically synchronized to the broadband seed pulses. Finally, the OPCPA must be designed in such way to avoid or suppress parametric superfluorescence (i.e. parametric amplification of vacuum fluctuations) in the nonlinear crystals.

CARRIER-ENVELOPE PHASE STABILIZATION WITH OPAS

The electric field of an ultrashort light pulse can be written as:

$$E(t) = Re[A(t)e^{i(\omega_0 t + \phi)}]$$

where A(t) is the complex field envelope, ω_0 is the carrier frequency and ϕ is the so-called carrier-envelope phase (CEP), which is the phase of the carrier wave with respect to the peak of the envelope. When the light pulse duration is shortened to a few optical cycles, the CEP becomes important, especially in highly nonlinear experiments such as high harmonic generation. CEP stabilization is at the heart of attosecond pulse generation, which was awarded with the Physics Nobel Prize in 2023. Within the cavity of a mode-locked oscillator, the carrier wave propagates with the phase velocity v_p, while the envelope propagates with the group velocity v_g; therefore, the CEP varies between consecutive pulses. Active approaches to CEP stabilization, which involve complex electronic circuits, have been demonstrated and are currently commercially available.

OPAs, on the other hand, offer the enticing opportunity to achieve fully passive all-optical CEP stabilization [6]. **Passive CEP stabilization is based on DFG between two pulses at frequencies ω_1 and ω_2 sharing the same CEP**, $\phi_1 = \phi_0 + c_1$ and $\phi_2 = \phi_0 + c_2$, where ϕ_0 is the shared CEP (fluctuating from pulse to pulse) and c_1 and c_2 are constant in time. It can be shown that the DFG pulse, at frequency $\omega_{DF} = \omega_2 - \omega_1$, carries the CEP $\phi_{DF} = \phi_2 - \phi_1 = c_2 - c_1$, which is constant, thus cancelling the CEP fluctuations of the interacting pulses. CEP stabilization naturally takes place in an OPA, in which the idler is generated by DFG between the pump and signal pulses, provided that the pump and the signal share the same CEP. This happens for example in an OPA pumped by the FF(SH) of a laser and in which the seed is generated by the FF(SH) of the laser. Such process in called interpulse CEP stabilization and is sketched in Figure 4. Alternatively, DFG can occur between different frequency components of a broadband pulse, in what is called intrapulse CEP stabilization.

With respect to active approaches, passive CEP stabilization has the advantages of the absence of electronic circuitry, of frequency tunability and of the possibility to combine ultrabroad bandwidths with energy scaling, using OPCPAs. In view of applications, however, the stability of active *vs.* passive CEP stabilization approaches needs to be fully • • •

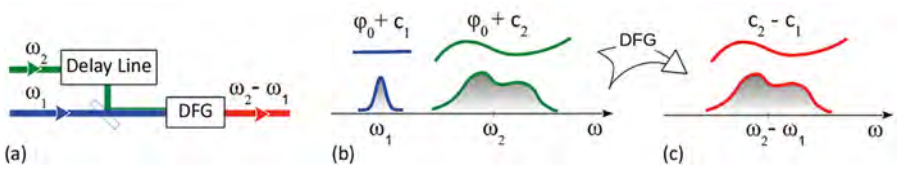


Figure 4. Interpulse CEP stabilization by the DFG/OPA process (a). (b) Spectral intensity and phase of each pulse. (c) The pulse at the DF; the common fluctuating spectral phase φ_0 has been cancelled by the nonlinear DFG process.

characterized and slow CEP drifts due to mechanical fluctuations should be compensated by suitable feedback loops.

COHERENT SYNTHESIS OF OPAS

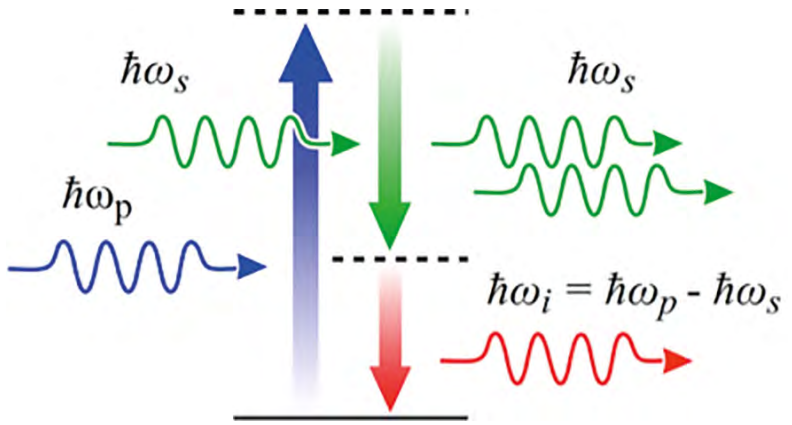
Group velocity matched OPAs or OPCPAs can generate pulses with bandwidth approaching one octave (i.e. containing a frequency f as well as its second harmonic 2f). Dispersion compensation then allows to compress these pulses down to a TL duration of 2-3 cycles of the carrier wave. Extending even more the bandwidth to cover multiple octaves would be appealing for many applications, especially in the so-called "extreme nonlinear optics" in which light-matter interactions under very high light field strengths are studied. However, achieving multi-octave gain bandwidths with OP(CP)As and accurate dispersion control over such a wide frequency range is very challenging. One way to further extend the bandwidth is to **use a *"divide et impera"* approach, in which the multi-octave spectrum is divided into multiple channels, each of which is separately amplified and compressed to TL duration** [7]. The channels are then coherently combined into a single ultra-broadband pulse. Coherent synthesis requires that the individual channels are synchronized and that their relative phases are locked using feedback loops based on balanced cross-correlators. In combination with CEP control, coherent synthesis allows to shorten the pulse width to the sub-cycle limit and generate high-power light transients with controlled electric fields, opening the research field of "waveform nonlinear optics".

Recently, we have demonstrated coherent synthesis of high-energy OPAs, generating phase-controlled sub-cycle light waveforms at the mJ energy level with excellent stability [8]. Full control over the synthesized waveforms, whose spectrum spans over 1.7 octaves with pulse duration down to 2.8 fs (corresponding to 0.6 optical cycles at a 1.4-µm central wavelength), has enabled the creation of extreme ultraviolet isolated attosecond pulses *via* high-harmonic generation without the need for additional gating techniques.

CONCLUSIONS

Nonlinear frequency conversion through OPAs greatly enhances the capabilities of ultrafast laser systems, enabling a plethora of applications in time-resolved spectroscopy, high-field science, materials processing, and biomedicine. The OPA technology is nowadays mature and commercially available; there are, however, several outstanding scientific and technological challenges. For day-to-day operation, robust and user-friendly architectures should be developed. For high sensitivity ultrafast spectroscopy applications, one should increase the repetition rate up to a few MHz and achieve shot-noise-limited operation.

OPTICAL PARAMETRIC AMPLIFICATION



Optical parametric amplification (OPA) is a **nonlinear parametric process because it proceeds via virtual states**, since light interacts with matter in such a way as to leave the final state of matter unchanged, without depositing any energy in the nonlinear material. It can be visualized as stimulated emission of signal photons with energy $\hbar\omega_s$ from a virtual level excited by the pump photons with energy $\hbar\omega_p$. The stimulated signal photons are in phase with the input ones, thus leading to constructive interference and hence to amplification. The process is completed by the emission of idler photons with energy $\hbar\omega_i$ for energy conservation, according to the formula: $\hbar\omega_p = \hbar\omega_s + \hbar\omega_i$. For the OPA process to be efficient, the so-called phase **matching condition** $k_p = k_s + k_i$, which guarantees that all the interacting waves are in phase, must be satisfied. The phase-matching condition corresponds to $\hbar k_p = \hbar k_s + \hbar k_i$, which is momentum conservation.

For high field science applications, the output energy should be increased up to tens of mJ and high CEP stability should be achieved.

Extension of frequency tunability also requires further research. The MIR wavelength range (3-10 µm) is important for time-resolved vibrational spectroscopy, for high harmonic generation, due to the λ_2 scaling of the cutoff energy [9], and for biomedical imaging (such as *e.g.* in photothermal IR spectroscopy). MIR pulses are currently produced by DFG between the signal and idler pulses of an OPA, with limited energy and bandwidth. Crystals with extended IR transparency typically have small bandgaps, with absorption onset already in the visible. Therefore MIR OPAs cannot be directly pumped by Ti:sapphire lasers, because of two-photon absorption of the pump pulse in the OPA crystal. The development of powerful ultrashort sources at ≈2 µm, based on Ho: and/or Tm:doped gain media, will allow direct pumping of MIR OPAs based on $ZnGeP_2$, GaSe and $AgGaSe_2$ crystals, with lower quantum defects and more favourable group-velocity matching. Ultrashort UV pulses are important for applications since many biomolecules (DNA, aromatic amino acids, protein cofactors) absorb in that wavelength range. UV pumping of OPAs is prevented by two-photon absorption of the pump pulse. SHG of visible pulses is limited by group velocity mismatch, unless achromatic phase matching is used. Alternatively, powerful UV pulses can be generated by SPM, exploiting resonant dispersive wave generation in a photonic crystal fiber.

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