Femtosecond optical parametric amplifiers with collinear phase matching: experiments and full simulation

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ABSTRACT

We report a high-power, kilohertz, ultrafast optical parametric amplifier (OPA) that is seeded by white-light continuum and contains three amplification stages. Two 3-mm KTA crystals cut in type-II phase-matching configuration are used in the OPA system which is capable of producing up to 70 µJ, 140 fs infrared laser pulses at wavelength ranging from 2.9 to 4 µm. A full-scaled numerical simulation on the OPA system was performed. Actual white-light seeded signal pulse and finite phase-matching bandwidth were taken into account in the calculation. Material dispersion and linear absorption of all the optical components involved were properly incorporated. The simulation results match the experimental results almost perfectly. Our simulation provides an essential tool to design and optimize the OPA systems. A step-by-step design procedure based on this simulation algorithm is presented.

Keywords: Femtosecond, optical parametric amplification.

1. INTRODUCTION

A high-power widely-tunable mid-infrared femtosecond laser source is essential in the ultrafast studies of inter-subband carrier dynamics in semiconductors and vibrational dynamics in chemical and biological systems. In the latter case, the fundamental stretching frequencies for O-H, N-H, C-H, and S-H bonds are of uttermost importance, thus requiring laser pulses that are tunable between 3 and 5 µm. The most popular method to generate infrared femtosecond light pulses is optical parametric generation, in which the photon energy of the pump wave is split into two lower-energy photons (signal and idler waves). A phase-matching condition derived from the momentum conservation relation is used to tune the output wavelengths of the signal and idler waves. Among many configurations of optical parametric generation, optical parametric amplification (OPA) with a collinear phase-matching configuration possesses the advantage of generating high-power nearly transform-limited ultrashort laser pulses with extensive wavelength tuning range. This configuration has thus been popularly utilized in ultrafast laser spectroscopy.

Although there have been mass theoretical studies, the understanding of this ultrafast optical technology is still incomplete. Also due to the limited understanding of OPA, it is difficult to determine the optimal design parameters before constructing the setup. For a long time, the design of ultrafast OPA systems had relied mostly on simple calculations (phase-matching condition, group-velocity mismatch, and output energy estimation) and try-and-error. To design an ultrafast OPA system, the designers need to answer the following questions: What are the requirements for the pulse duration and the energy of the pump pulse? What is the nonlinear optical crystal to be used with the available pump laser wavelength? What is its thickness? What is the phase-matching type? How many amplification stages are needed? What are the pump laser intensities at different stages? It is a difficult task for the designers to finalize these parameters from the required output specifications. Here we report a complete numerical simulation of collinear phase-matching femtosecond optical parametric amplifier seeded by white-light continuum. Like in previous theoretical efforts, group-velocity mismatch, dispersion, and linear absorption are properly incorporated. Moreover, in our efforts, actual white-light seeded signal pulses and finite phase-matching bandwidth are considered. The validity of our simulation was verified by our actual OPA system. A design procedure based on the simulation algorithm was developed.

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2. EXPERIMENT

The femtosecond pump source consists of a mode-locked Ti:sapphire laser (Tsunami, Spectra-Physics Lasers) pumped by a CW diode-pumped Nd:YVO₄ laser (Millenia, Spectra-Physics Lasers), and a Ti:sapphire regenerative amplifier system (Super-Spitfire, Spectra-Physics Lasers) pumped by the second harmonics of two CW Q-switched Nd:YLF lasers (Merlin, Spectra-Physics Lasers). The resultant output consists of 1 kHz, 3 mJ, and ca. 140 fs pulses with wavelength centered at 800 nm. 90% of the output energy is used to pump the OPA. The remaining is used to characterize the idler pulse duration in a cross-correlation experiment. The pump beam is further split with a 50/50 beamsplitter. Figure 1 depicts a schematic diagram of the home-build OPA system. It consists of three amplification stages and two 3-mm KTiOAsO₄ (KTA) crystals. A small portion of the first pump beam (95/5 beamsplitter, BS1) was focused onto a 2-mm sapphire disk to generate white-light seeded signal pulses (white-light generator), which was then directed via a delay line (D1) onto the first KTA crystal (KTA1). The combination of a zero-order half-wave plate (λ/2) and a thin film polarizer (TFP) was used to finely tune the input pulse energy in white-light generation. An iris diaphragm (ID) was used to further adjust the self-focusing condition in the sapphire disk. The major portion of the first pump beam was split into two beams by a 15/85 beamsplitter (BS2). The smaller reflected portion was made collinear with the white-light seeded signal pulse and directed by a dichroic mirror (DBS1) onto KTA1 for the first amplification stage. The generated IR waves was reflected off a curved Ag mirror (CM) to become collimated and then directed onto KTA1 but slightly vertically displaced. The residual 800 nm pump was removed by a dichroic mirror (DBS2). The larger portion of the first pump beam was combined with the reflected IR using the same dichroic mirror (DBS2). The temporal overlap was controlled by the delay line D2. This is the second amplification stage. The residual 800 nm pump was removed by a dichroic mirror (DBS3) and the IR output (signal plus idler) of the second amplification stage was directed onto the second KTA crystal (KTA2), where it was made collinearly overlapped with the second pump beam. The temporal overlap was controlled by the delay line D3. After this third amplification stage, the residual 800 nm pump beam and most of the signal beam were removed by DBS5 and DBS6, respectively. The two KTA crystals are cut for type-II phase matching at θ = 43º and φ = 0º. Light waves propagate in the xz plane. The signal wave (extraordinary wave) is polarized in the xz plane. Both the pump and idler waves (ordinary waves) are polarized along the y axis. The two crystal surfaces are coated with anti-reflection coating at 800 nm to minimize the reflection loss of the pump beam.

The average power of the OPA IR output was measured by a thermopile detector (PM3, Molectron). The wavelengths and spectral widths were measured by a 30-cm monochrometer (SpectraPro 300i, Acton Research Corp.) with a 150-grooves/mm grating blazed at 4 μm, in combination with a liquid-nitrogen cooled InSb detector. A small portion of the amplifier output was used to characterize the idler pulse duration. The reflection sum-frequency generation

![Figure 1. Layout of the optical parametric amplifier system. PS is a periscope, BS1 and BS2 are beamsplitters (R/T = 95/5 and 15/85, respectively), ID is an iris diaphragm, WR is a wave plate, TFP is a thin film polarizer, CM is a curved mirror, and DBS1~DBS6 are dichroic mirrors. D1~D3 are delay lines. The white light generator consists of two convex lenses and a 2-mm sapphire plate.](image)
signal, from a GaAs wafer, produced by the 800-nm amplifier output and the idler beam was used to obtain cross-correlation traces. The autocorrelation trace of the amplifier output pulses was also recorded. Assuming Gaussian pulse shape for both the amplifier output and the idler pulse, the idler pulse duration was retrieved from the cross-correlation trace and the autocorrelation trace. This OPA system is capable of producing 1-kHz ~150 fs mid-infrared laser pulses at wavelengths ranging from 2.9 to 4.0 µm with energy as high as 70 µJ.

Although using a white-light seeded signal pulse is very popular in many home-made and commercial OPA systems, the actual field amplitude and phase of this white-light seeded pulse has, to our knowledge, never been incorporated in the simulation. Instead of speculating the white-light continuum based on existing theories,9,10 we have made a tremendous effort to characterize it. The experimental characterization of the white-light continuum was carried out between 1 and 1.1 µm that is the seeded signal spectral range corresponding to the idler wave tunable from 2.9 to 4.0 µm. The white-light continuum was first dispersed by a 1/4-m monochrometer (Oriel Instruments) and detected by a calibrated thermoelectric-cooled InGaAs photodiode. The spectral energy density of the white-light continuum was then determined. An artificially generated band-pass filter was used to select the seeded signal wavelength range corresponding to the tuning range of the real OPA system. To obtain the temporal information, the white-light beam and a small portion of the 800-nm amplifier output beam were focused non-collinearly into a 1-mm β-BaB₂O₄ (BBO) crystal (Type-I phase matching) to produce sum-frequency generation signals. The cross-correlation traces were then recorded over the relevant seeded signal wavelength range. Based on the final energy spectrum and the cross-correlation traces, a cross-correlation frequency-resolved optical gating (XFROG) trace was constructed. The field amplitude and phase of the white-light seeded signal pulse were then retrieved with a phase-retrieval algorithm (FROG 3.08, Femtosoft Technologies). The filter bandwidth used to truncate the power spectrum was adjusted until it covers the whole signal wavelength range of interest and reaches a spectral bandwidth that a wider spectral range produces only minor variation in the retrieved field amplitude and phase.

3. NUMERICAL SIMULATION

Assuming slowly-varying envelope approximation and infinite plane wave, the second-order nonlinear coupling equations that govern the nonlinear interaction of the pump, signal, and idler waves can be derived directly from the Maxwell equations.1 Interactions between the three waves were described by the second-order nonlinear polarizations, \( \{ P_{NL} \} \), which, to account for the phase matching condition, are expressed as follows:

\[
P_{NL}^{P}(z,\omega) = \varepsilon_o \chi_{eff}^{(2)} \left[ \int d\omega' F_s(\omega') F_i(\omega - \omega') \right] \exp[-ik_p(\omega)\Delta z],
\]

\[
P_{NL}^{S}(z,\omega) = \varepsilon_o \chi_{eff}^{(2)} \left[ \int d\omega' F_p(\omega') F_i(\omega - \omega') \right] \exp[-ik_s(\omega)\Delta z],
\]

and

\[
P_{NL}^{I}(z,\omega) = \varepsilon_o \chi_{eff}^{(2)} \left[ \int d\omega' F_p(\omega') F_s(\omega - \omega') \right] \exp[-ik_i(\omega)\Delta z],
\]

where \( F_j(z,\omega) = B_j(z,\omega) \exp[ik_j(\omega)\Delta z] \). \( B_j(z,\omega) \) is the Fourier transform of the time domain field envelope, and \( \Delta z \) is the increment of propagation distance. These derivations are based on the fact that the finite spectral width of ultrashort laser pulses should be considered when calculating the nonlinear polarizations. The output wave is the sum of all the generated frequency components that satisfy the energy conservation: \( \omega_p = \omega_s + \omega_i \). Accordingly, there are many such frequency combinations to generate one specific frequency. These wavelength combinations should have different gains. Nonzero phase mismatch therefore needs to be taken into account. At each propagating increment, the nonlinear polarizations for the pump, signal, and idler waves were calculated based on the electric fields at the previous step. With material dispersion and linear absorption, the nonlinear coupled wave equations were then solved numerically by the fourth-order Runge-Kutta method.1,11

The electric fields of the pump pulse and the white-light seeded pulse were prepared first. The pump pulse is assumed to be a linearly chirped Gaussian pulse. Its characteristics, i.e. pulse duration, spectral width, and chirp parameter were experimentally determined. In the actual operation of the OPA system, the three delay lines (D1, D2, and D3)
are adjusted to obtain maximum output energies for the first, second, and third amplification stages, respectively. Our simulation emulates these actions by adjusting the temporal positions of the pump pulse to maximize the output energy for each amplification stage. The material dispersion and the linear absorption of the KTA crystal and the dichroic beamsplitters in the OPA system (DBS2-DBS6) were incorporated in the calculation.

For a specific output idler wavelength, the corresponding signal wavelength was first calculated with the energy conservation relation. The phase-matching angle of the desired output idler wavelength was then determined. Subsequently, the dispersion relation of each wave was computed based on the birefringence relation of the KTA crystal. With the pre-determined phase-matching angle, every coefficient in the nonlinear coupling equations was determined accordingly. The output idler wavelength is a natural outcome of the phase-matching consideration, Eqs. (1)-(3), from the simulation. The simulation was done at the output idler wavelength of 3 $\mu$m. The beam diameter of the seeded signal beam and the pump energy density were slightly adjusted around their experimentally determined values to match the simulated output pulse energy of the idler pulse with the experimental value at each amplification stage, while the pulse shape of the idler pulse was monitored simultaneously to make a good correspondence with the cross-correlation measurement. After this adjusting process, the simulation was then performed on other idler wavelengths without any further adjustment in the pump energy density and the seeded signal beam diameter. No artificial scaling factor was needed to match the experimental results.

4. RESULTS AND DISCUSSION

The result of extracting the field amplitude and phase of the white-light seeded signal pulse was presented first. Figure 2 shows the constructed and retrieved XFROG traces of the white-light seeded signal pulse. Figure 3 shows the retrieved intensity and phase as a function of time delay and the intensity spectrum. Self-steepening and pulse shortening phenomena appear in the pulse shape, indicating the occurrence of self-focusing during the supercontinuum generation process. The power spectrum decays exponentially in the spectral range of interest (from 1 to 1.1 $\mu$m) and is consistent with our measured result. The cut-offs at both 0.98 and 1.15 $\mu$m are a consequence of the artificially imposed truncated filter. The retrieved results are considered to be correct because no significant difference was discovered when enlarging the spectral bandwidth of this truncated filter.

Figure 2. (a) Constructed and (b) retrieved FROG traces of the white-light continuum.

Figure 4(a) shows both the experimental and simulation results of the idler output energy as a function of wavelength. From 2.9 to 4.0 $\mu$m, the output power drops slightly from 70 $\mu$J to about 60 $\mu$J, corresponding to a photon conversion efficiency varying from 10.4% to 11.3%. The simulation results match the experimental data very well. The energy drop at 2.9 $\mu$m is due to the absorption by water vapor in this spectral region, which is not taken into account in our simulation. This wavelength dependence of the idler output pulse energy is roughly consistent with the experimental results by Gragson et al., Petrov et al., and Cussat-Blanc et al.
Figure 3. (a) Retrieved intensity (solid line) and phase (dashed line) of the white-light seeded signal pulse. (b) Intensity spectrum of the white light seeded signal.

Figure 4. Experimental and simulation results of the OPA system: (a) Output energy, (b) pulse duration (FWHM), (c) spectral width (FWHM), and (d) time-bandwidth product (TBP). The black dots are the experimental data. The simulation results are connected by solid lines.
Figure 4(b) shows the pulse duration (FWHM) of the idler wave retrieved from the cross correlation measurements assuming Gaussian pulse shape. The simulation results follow the trend of the experimental data very well. The wavelength dependence of the spectral width (FWHM) is shown in Fig. 4(c). The spectral spectrum is also assumed to be Gaussian shape. From 3.0 to 3.8 µm, the spectral width increases with the idler wavelength from 83 to 123 cm⁻¹, agreeing roughly with the simulation results. According to the results in Figs. 4(b) and (c), the time-bandwidth product was calculated and is shown in Fig. 4(d). The value increases slightly with the idler wavelength and is very close to the transform limit. The good match between the experimental and the simulation results indicates that our simulation program can account for almost every practical aspect in the OPA system.

Figure 5 shows the pulse duration (FWHM) and photon density of the pump, signal, and idler waves as they propagate inside the KTA crystal of the first amplification stage. The sudden drop and abrupt rise in Fig. 5 (a) are due to pulse splitting. According to the results shown in Fig. 5, we can divide the amplification process into three regions: the pulse shaping (PS) region, stable amplification (SA) region, and gain saturation (GS) region. In the PS region, the signal wave has gone through a shaping process. The pump pulse maintains its pulse shape since no significant energy transfer takes place. After the PS region, the signal and idler pulse reach stable pulse shape. Both the signal and idler waves experience linear-gain amplification and sustain stable pulse shape in the SA region. The photon densities of the signal and idler waves merge gradually in the SA region. The location where these two photon densities finally meet marks the end of the influence of the seeded signal pulse. In the GS region, the energy flows back and forth between the pump, signal, and idler waves and the pulse shape of the three waves distorted and split. From a practical point of view, only pulses under SA region possess integral pulse shape that is suitable for subsequent applications. The thickness of the nonlinear crystal should be chosen such that the OPA process stops within the SA region. In contrast, simple estimation based on GVM and GVD was adopted in previous OPA design, which is insufficient for resolving this problem.  

Figure 5. (a) Pulse duration (FWHM) and (b) photon density of the pump (dotted line), signal (dashed line), and idler (solid line) waves as they propagate inside the KTA crystal.

5. DESIGN PROCEDURE

From the discussion above, we know that OPA is a highly nonlinear process. Simple estimation (usually based on linear calculation) cannot predict the output characteristics as well as the optimum design parameters. The almost perfect match between the experimental and the simulation results described here proves that our simulation provides an essential tool to design and optimize OPA systems. The design concept of such a complicated device is not as simple as providing a solution to a question. It should be similar to the case of designing a complicated integrated circuit. That is, our simulation algorithm provides instantaneous feedback to an OPA design parameter set (the design phase space) to check whether the output specifications are met. The design parameters can then be adjusted and are used again for the simulation. This procedure is preceded iteratively and the parameters can then gradually move to the optimal point in the design phase space. Our simulation algorithm serves as a virtual OPA system to test on the design.
parameters without physically constructing one. An illustrative step-by-step design procedure is explained in the following.

*Step 1 (Identifying the output specifications):* In the first step, the output pulse duration, energy and wavelength tunable range are specified.

*Step 2 (Choosing the pump laser source):* The output pulse duration follows roughly with the pulse duration of the pumping laser system. Because the conversion efficiency in the OPA process is normally 10%, the pump pulse energy should be estimated accordingly.

*Step 3 (Choosing the nonlinear crystal and the phase-matching type):* The nonlinear optical crystal needs to be transparent and the phase-matching condition needs to be satisfied throughout the specified wavelength tunable range. The spectral bandwidth, the conversion efficiency and the ease of separating signal and idler waves are also taken into account in determining the appropriate phase-matching type.

*Step 4 (Characterizing the pump pulse):* The field amplitude and phase of the pump beam should be characterized. Presumably, a FROG measurement can be performed on the pump pulse to extract the exact field amplitude and phase.

*Step 5 (Characterizing the white-light seeded signal pulse):* The field amplitude and phase of the white-light seeded signal pulse should be retrieved from the XFROG trace by a phase-retrieval algorithm. The white-light characteristics depend on the nonlinear optical material, its thickness, the pump laser characteristics, and the focusing condition etc.

*Step 6 (Estimating the seeded energy density):* In order to boost up the amplification gain in the first amplification stage, the white-light seeded signal beam is usually focused at the nonlinear crystal. Because of material dispersion, it cannot be focused tightly. The re-collimating lens after nonlinear optical material should be selected properly to ensure the depth of focus being longer than the crystal thickness. This requirement comes from the plane-wave propagation assumption used in our simulation and thus provides a criterion to determine the beam size of the white-light seeded signal beam. After measuring the diameter at the focal point, the energy density of the white-light seeded signal pulse can be calculated accordingly.

*Step 7 (Guessing the pump energy density):* In the beginning of the iterative process, an initial guess of the first-stage pump energy density is used. Its value can be adjusted in the later iterative steps. The upper bound is the threshold of spontaneous parametric fluorescence and the lower bound is the energy density of the white-light seeded signal pulse. The beam size of the pump beam needs to be larger than that of the white-light seeded signal beam. With the pump beam size determined, the pump energy density can be calculated accordingly.

*Step 8 (Simulating the first amplification stage):* The program is run until the gain saturation region is reached. The temporal delay of the pump pulse is adjusted to maximize the output energy.

*Step 9 (Does the output pulse quality meet the output specification?):* The output energy of the first amplification stage is a secondary concern. The first amplification stage should end at a point in the SA region where good pulse quality is produced and allows for calculating error and pump energy density adjustment. If the resultant pulse quality does not meet the specified requirements, further adjustment of the focusing condition of the pump and white-light seeded signal beams in *Step 10* is needed. Otherwise, the next step is *Step 11*.

*Step 10 (Adjusting the pump and seeded energy density):* If the pulse quality (pulse duration and time-bandwidth product) does not meet the specifications, either or both the focusing condition of the pump and white-light seeded signal beams should be readjusted. The procedure goes back to *Step 8*.

*Step 11 (Determining the crystal thickness of the first amplification stage):* The crystal thickness should be chosen such that the amplification ends at a point in the SA region in which high enough output energy is produced while maintaining good pulse quality.

*Step 12 (Does the output energy meet the output specification?):* If the output energy is not large enough, subsequent amplification stages are then needed.
Step 13 (Propagating pulses through the optical components between amplification stages): In normal OPA systems, dichroic beamsplitters are often used to steer the pump beam to be collinear with the unamplified beam. The pulse duration of the signal and idler waves is broadened and the temporal relation of these two waves is varied, because of group-velocity dispersion and group-velocity mismatch induced by the material dispersion property of these optical components. Moreover, their additional linear loss reduces the energy of resultant signal and idler waves. These effects need to be taken into account by propagating both the signal and idler pulses through the optical material with an equivalent optical thickness.

Step 14 (Guessing the energy density of the signal and idler beam): Unlike the first amplification stage, the iterative process of the second amplification stage begins with an initial guess of both the energy density of the signal and idler beams as well as the second-stage pump energy density. To prevent further gain saturation, the energy density of the unamplified signal and idler beams should be reduced from that at the output of the first amplification stage by expanding their beam diameter. With an initial guess of the beam size, the energy density of the signal and idler beams for the second amplification stage can be calculated accordingly.

Step 15 (Guessing the pump energy density): The realistic limitations of the pump energy density are the available pump pulse energy, the threshold for optical parametric fluorescence, and the damage threshold. With the pump beam size determined, the pump energy density can be calculated accordingly.

Step 16 (Simulating the second amplification stage): The program is then run until the gain saturation region is reached. The temporal delay of the pump pulse is adjusted to maximize the output energy.

Step 17 (Dose the output pulse quality meet the specification?): Similarly, the amplification should end at a point in the SA region where allows for calculating error and pump energy density adjustment. If the resultant pulse quality does not meet the specification, a further adjustment of the energy density of the pump, signal, and idler waves in Step 18 is needed. Otherwise, the procedure proceeds to Step 19.

Step 18 (Adjusting the pump, signal, and idler energy density): If the pulse quality obtained in Step 17 does not meet the specified specification, either or all the energy density of the pump, signal, and idler beams should be readjusted. Then the next step is Step 17 with new parameters.

Step 19 (Determining the crystal thickness of the subsequent amplification stage): The crystal thickness should be chosen such that the amplification ends at a point in the SA region in which high enough output energy is produced while maintaining good pulse quality.

Step 20 (Dose the output energy meet the specification?): If the output energy is not large enough, subsequent amplification stages are then needed. The procedure goes to Step 13.

6. CONCLUSIONS

We have succeeded in generating very intense pulses (up to 70 µJ) between 2.9 and 4 µm, with time-bandwidth product values close to transform limit, using a three-stage OPA design. A numerical simulation algorithm has been developed based on this system. In this simulation, the actual field amplitude and phase of the white-light seeded signal pulse has been incorporated in the calculation and phase-matching condition has been properly accounted for. The almost perfect match between the experimental and the simulation results described here proves that our simulation provides an essential tool to investigate and design the OPA system. Three amplification phases, for the first time, has been recognized: the pulse shaping, steady amplification, and gain saturation. This discovery has made the analysis easier. Finally, in designing an optimal ultrafast OPA system, our simulation algorithm provides instantaneous feedback in the iterative design process.

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