Compact dual-crystal optical parametric amplification for broadband IR pulse generation using a collinear geometry

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Abstract: A novel compact dual-crystal optical parametric amplification (DOPA) scheme, collinearly pumped by a Ti:sapphire laser (0.8 μm), is theoretically investigated for efficiently generating broadband IR pulses at non-degenerate wavelengths (1.2 μm~1.4 μm and 1.8 μm~2.1 μm). By inserting a pair of barium fluoride (BaF2) wedges between two thin β-barium borate (BBO) crystals, the group velocity mismatch (GVM) between the three interacting pulses can be compensated simultaneously. In this case, the obtained signal spectrum centered at 1.3 μm is nearly 20% broader and the conversion efficiency is increased, but also the pulse contrast and beam quality are improved due to the better temporal overlap. Furthermore, sub-two-cycle idler pulses with carrier-envelope phase (CEP) fluctuation of sub-100-mrad root mean square (RMS) can be generated. Because a tunable few-cycle IR pulse with millijoule energy is attainable in this scheme, it will contribute to ultrafast community and be particularly useful as a driving or controlling field for the generation of ultrafast coherent x-ray supercontinuum.

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References and links
1. Introduction

High-order harmonic generation (HHG) driving by femtosecond laser pulses has attracted a lot of attention for producing fully coherent radiation in the extreme ultraviolet (XUV) region, which may serve as a table-top alternative to the x-ray lasers [1]. It has also opened the door of attosecond science, in which the shortest pulse duration of isolated attosecond pulse (IAP) attained is 67 as [2]. However, the generation of IAPs of duration down to the order of 100 as or less with few-cycle pumping [3], polarization gating [4] and double optical gating (DOG) [2] schemes, essentially requires a few-cycle (< 7 fs) driving laser and CEP stabilization. To produce an IAP with a longer driving pulse and suppress the ionization for scaling up the IAP yields, an optimized two-color synthesis, composed of a 30-fs/0.8-μm Ti:sapphire main field and a 40-fs/1.3-μm assistant field, was recently shown by Takahashi and Lan et al. [5, 6]. Although the used pulse durations are available from commercial laser amplifiers, the duration of the generated IAP is sub-femtosecond, which is not sufficient for a number of fundamental processes such as the intra-atomic energy transfer between electrons, the response of electron system to external influence and so forth [3, 7, 8]. Consequently, sub-15-fs two-color synthesis is highly desired for generation of a 100-as IAP, as previously reported in [9, 10]. This makes the generation of sub-15-fs intense laser source around 1.3 μm urgent and important.

To further develop the applications of IAPs, the other important issue is the extension of the central wavelength into the soft-x-ray region. It has realized that the maximum harmonic photon energy by $E_{\text{cutoff}} = I_p + 3.17 U_p$, where $I_p$ is the ionization potential of the target atom, $U_p$ [eV] $= 9.38 \times 10^{-14} I \left[\text{W/cm}^2\right] (\lambda \left[\mu\text{m}\right])^2$ is the ponderomotive energy of the electron in the
laser field. Since the $U_p$ is proportional to the $\lambda^2$, one effective approach to significantly extend the harmonic cutoff is to adopt a longer-wavelength driving field. Using this concept, Takahashi et al. have succeeded in generating harmonics with photon energies of 300 eV and 450 eV [11, 12]. By further increasing the driving wavelength to $\sim$4 $\mu$m, Popmintchev et al. have obtained ultrahigh harmonics in the keV x-ray regime [13]. Yakovlev et al. have theoretically shown that the IAPs can be extracted from harmonic spectrum using CEP-stabilized few-cycle 2.1-$\mu$m driving source [14]. In order to generate sub-100-as IAPs in soft-x-ray region, a two-color synthesis, combing a 12.5-fs/2.0-$\mu$m driving pulse and a weaker 0.8-$\mu$m controlling field, was recently proposed by Hong et al. [15]. These pioneering experimental results and theoretical predictions depict a bright prospect for soft-x-ray IAPs, and therefore simulate the quest for sub-10-fs intense laser sources with central wavelengths around, for example, 2.0 $\mu$m and so on.

Due to the lack of laser material, the CPA technology cannot be applied in $>1$ $\mu$m spectral region. An alternative optical parametric amplification (OPA) has become the most promising way to down-convert Ti:sapphire laser light for its advantages including broad gain bandwidth, less accumulation of nonlinear effects, a low thermal load on nonlinear crystal and the tunability in gain spectral region. Based on 0.8-$\mu$m laser pumped OPAs, near-infrared (NIR) to mid-infrared (MIR) femtosecond pulses have been widely realized [16–20]. A major concern in ultrabroadband, high-efficiency OPA designs is the GVM between three interacting pulses. The temporal walk-off induced by GVM can severely limit the bandwidth of the output spectrum and the effective interaction length [21].

Since the phase-matching condition for spectral acceptance is limited by the GVM between signal and idler (GVM$_{si}$), to broaden the gain spectrum for few-cycle pulse generation, degenerate OPAs have been proposed and realized in many previous works [22–25]. Owing to the identical signal and idler wavelengths at degeneracy, phase matching and group velocity matching are simultaneously satisfied, therefore ultrabroad gain bandwidth can be expected [16]. With 0.8-$\mu$m pump, Brida et al. [23] achieved broadband amplification of degenerate 1.6-$\mu$m signal, and sub-two-cycle (8.5 fs) near-IR pulses were obtained. Even though degenerate OPA is a promising method to generate few-cycle pulses, it is restrained at a particular wavelength with poor tunability. For non-degenerate seed (1.2 $\mu$m $\sim$ 1.4 $\mu$m), broadband amplification faces technical challenges due to the large GVM$_{si}$. By introducing an initial pump-signal noncollinear angle $\alpha$ so that the signal group velocity equals the projection of the idler group velocity along the signal direction, the amplified pulses (signal and idler) can be overlapped over a wide range, allowing broader gain bandwidth at non-degenerate wavelength region [26–30]. In Ref. [30], the noncollinear OPA (NOPA) system generated IR pulses with a spectrum covering 1.1 $\mu$m to 1.7 $\mu$m, which supports sub-10-fs pulse duration. However, this noncollinear geometry results in angularly dispersed idler, consequently complex and energy-consuming angular dispersion compensating stage is necessary to obtain a collimated idler beam for its further use [31, 32].

Apart from the compensation of GVM$_{si}$ for a broader gain bandwidth, it is also of importance to compensate the GVM between the pump and signal/idler (GVM$_{ps}$ and GVM$_{pi}$) to increase the conversion efficiency in OPA, particularly for ultrashort interacting pulses. The GVM$_{ps}$ and GVM$_{pi}$ will lead to spatially and temporally pulse splitting, which restricts the effective interaction length in the nonlinear crystal. To match the group velocities of three pulses in the crystal, the pulse-front matching scheme has been proposed [31, 33, 34]. This configuration can efficiently enhance the three-pulse overlap and eliminate the pulse-front tilting effect, thus improving the gain efficiency during interaction. Nevertheless, tilting wave fronts is equivalent to introducing angular dispersion into the pulse, prisms or gratings are therefore required to recollimate the beams, adding the system complexity.

In this paper, we present a novel dual-crystal OPA (DOPA) scheme to compensate the three-
pulse GVM simultaneously by inserting a pair of BaF$_2$ wedges between two pieces of thin BBO crystals. A compact collinear geometry, which is free of angularly dispersed pulses, is employed for simplicity. Besides, the DOPA scheme is easy to achieve low-timing-jitter all-optical synchronization and implement, because the pump and signal pulses come from a common Ti:sapphire source. Numerical calculations of coupled-wave equations are carried out to investigate the spectral and temporal characteristics of the generated IR pulses. The results show that both the gain bandwidth and conversion efficiency are improved in this scheme, meanwhile the amplification of quantum noise and pulse side lobe can be markedly suppressed. Additionally, CEP-stabilized sub-two-cycle idler pulses are obtained in the MIR region. The rest of this paper is arranged as follows. In Sec. 2, we illustrate the effects of three-pulse GVM in OPA and introduce the concept of our compact DOPA scheme. In Sec. 3, we study the improvement of signal bandwidth and conversion efficiency by numerically simulating the OPA process. In Sec. 4, the superfluorescence and B-integral buildup in DOPA is investigated. In Sec. 5, we analyze the gain bandwidth and CEP fluctuation of the idler pulses. Finally in Sec. 6, the conclusions are drawn and prospects for DOPA development are discussed.

2. Group velocity mismatch in ultrafast OPA

First, we discuss the influence of GVM on OPA performances. During the OPA process, the seed pulse can only be efficiently amplified when both the law of energy conservation ($\hbar \omega_p = \hbar \omega_s + \hbar \omega_i$) and momentum conservation ($k_p = k_s + k_i$) are satisfied, here subscripts $p$, $s$, and $i$ stand for the pump, signal/seed and idler waves, respectively. To evaluate the performance of OPA process, an important parameter called phase mismatch (wave vector mismatch) $\Delta k$ is introduced. Assuming a monochromatic pump, the phase mismatch can be expanded as

$$\Delta k = \Delta k_0 + \left( -\frac{\partial k_s}{\partial \omega_s} \Delta \omega + \frac{\partial k_i}{\partial \omega_i} \Delta \omega \right) + \frac{1}{2!} \left( \frac{\partial^2 k_s}{\partial \omega_s^2} + \frac{\partial^2 k_i}{\partial \omega_i^2} \right) \Delta \omega^2 + \cdots$$

(1)

Usually, the OPA is designed to make sure that a perfect phase matching is fulfilled for the central wavelengths of pump, seed and idler pulses, i.e. $\Delta k_0 = k_p - k_s - k_i = 0$. Neglecting second and higher order dispersions, the phase mismatch can then be approximated to first order as

$$\Delta k_1 = \frac{\partial k_s}{\partial \omega_s} \Delta \omega + \frac{\partial k_i}{\partial \omega_i} \Delta \omega = -\left( \frac{1}{v_{gs}} - \frac{1}{v_{gi}} \right) \Delta \omega.$$  

(2)

It can be seen from Eq. (2) that $\Delta k_1$ is proportional to $1/v_{gs} - 1/v_{gi}$, namely GVM$_{si}$. Therefore, ultrabroadband gain can be realized in a collinear degenerate OPA when the signal and idler share the same group velocity. However, as the signal frequency goes away from degeneracy, the GVM$_{si}$ grows rapidly and the phase-mismatch increases, strongly narrowing the gain bandwidth in non-degenerate region.

Besides the gain bandwidth, the conversion efficiency is also a significant factor for OPA, which is determined by the GVM$_{ps}$ and GVM$_{pi}$. The effective interaction between the three pulses only takes place when they overlap temporally, the available crystal length is therefore limited by the three-pulse temporal walk-off, which largely depends on the propagating directions of signal and idler pulses relative to the pump [21]. If the signal and idler pulses travel away from the pump peak in the same direction, the pulses drift to the weak edge of the pump, thus the parametric gain saturates rapidly and high conversion efficiency cannot be achieved. On the other hand, if they walk off in opposite directions, the amplified pulses tend to stay localized under the pump and the gain increases exponentially with respect to the crystal length, even when it is longer than the pulse splitting length. We analyze a 0.8-\textmu m laser pumped OPA
using type-I \((o_s + o_i \rightarrow e_p)\) phase matching for example. Because of the prominent characteristics including broad phase-matching range (0.41 \(\mu m\) - 3.5 \(\mu m\)), wide transparency range (0.19 \(\mu m\) - 3.5 \(\mu m\)), large nonlinear coefficient, and high damage threshold, BBO crystal is employed. The GVM\(_{ps}\) and GVM\(_{pi}\) are defined as 

\[
\delta_{pj} = \frac{1}{v_{gj}} - \frac{1}{v_{gp}}, j = s, i.
\]  

In this case, \(\delta_{pj} > 0, j = s, i\), both the signal and idler pulses propagate slower than the pump during amplification and they walk off in the same direction. The parametric gain will remarkably decrease as the signal/idler walk away from the pump peak and overlap with the weak edge or even split from the pump completely.

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Fig. 1. Configuration of (a) SOPA and (b) DOPA.

To simultaneously compensate both the GVM\(_{si}\) and the GVM\(_{ps}/GVM_{pi}\) in type-I OPA, a compact dual-crystal configuration with a third crystal for GVM compensation is proposed. Figure 1 clarifies the difference between the single-crystal OPA (SOPA) and the DOPA. The configuration of SOPA is shown in Fig. 1(a). During the interaction, the three pulses with different group velocities walk away from each other temporally. In principle, the accumulated signal-idler walk-off can be suppressed using a thinner crystal, thus a larger gain bandwidth is preserved. However, the extracted signal energy decreases due to its shorter interacting length. To maintain the broad bandwidth of the thin crystal while increase its conversion efficiency, as Fig. 1(b) shows, a GVM-compensating crystal with low nonlinearity is inserted between two pieces of thin BBO crystals in our scheme. The GVM-compensating crystal is desired to have opposite group-velocity relationship \((v_{gp} < v_{gs} < v_{gi})\) to the BBO crystal \((v_{gp} > v_{gs} > v_{gi})\). Therefore, the three-pulse temporal overlap is significantly enhanced by the crystal and much higher gain efficiency can be expected in the second stage. As a result, both the gain bandwidth and conversion efficiency will be increased. Besides, to improve the tunability, the GVM-compensating crystal is shaped into a pair of wedges so that the crystal length can be adjustable.
for different signal wavelengths. Based on the foregoing illustration, several kinds of commercially available crystals are compared and BaF$_2$ turns out to be the most appropriate one for this use, therefore it is applied in our following simulations.

3. GVM-compensating effect on signal pulse

The parametric amplification process is simulated by numerically solving coupled-wave equations with split-step Fourier-transform algorithm [35–37], the nonlinear interaction, temporal walk-off and crystal surface reflection are included. We assume a Gaussian-type 40-fs pump at 0.8 $\mu$m with an energy of 3 mJ and a beam diameter of 10 mm, corresponding to a peak intensity of 100 GW/cm$^2$, which is below the damage threshold of BBO and BaF$_2$ crystals [38,39]. A broadband Gaussian-type seed supporting 10-fs transform-limited (TL) duration is used for the amplification. Crystal parameters in the SOPA and DOPA are shown in Table 1. The crystal angles are chosen to satisfy the type-I phase-matching conditions for different seed wavelengths. The BBO lengths in SOPA are selected to drive the seed to saturation so that the signal gets amplified sufficiently, meanwhile the pump-signal delay is optimized to 0 fs for the broadest output spectrum. The particular dual-BBO length is selected to ensure the same gain as in SOPA, then the bandwidth of these two schemes are compared. We adjust the BaF$_2$ crystal length to precisely compensate the three-pulse GVM so that the interacting pulses can be synchronized when they are extracted, as shown in Fig. 1(b). With the designed parameters, the time delay between the output pump and signal/idler is suppressed under 2 fs for all wavelengths, which is beneficial for improving the OPA performances. From an experimental point of view, the realizability of BaF$_2$ wedge pair faces technological challenges due to its large aperture and small wedge angle, customer-desired trapezoid-shaped BaF$_2$ pair can be fabricated with an aperture of 20 $\times$ 20 mm$^2$, the thicknesses of two edges are 0.1 mm and 0.7 mm, respectively. With this BaF$_2$ pair, the crystal thickness can be tunable from 0.5 mm to 1.1 mm for $\phi$ = 10 mm beam. To compensate the GVM of interacting pulses with a larger beam size, there is no realizable BaF$_2$ wedges that can be applied. In this case, a flat-shaped BaF$_2$ crystal with a specific thickness will be used, e.g., for the 1.2-$\mu$m signal pulse with a 20-mm diameter, the BaF$_2$ crystal with a size of $\phi$ 25 mm $\times$ 1.4 mm can be applied.

<table>
<thead>
<tr>
<th>$\lambda_s$ ($\mu$m)</th>
<th>Crystal angle (°)</th>
<th>Single-BBO length (mm)</th>
<th>Dual-BBO length (mm)</th>
<th>BaF$_2$ length (mm)</th>
</tr>
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<tbody>
<tr>
<td>1.2</td>
<td>20.28</td>
<td>2.2</td>
<td>0.8 + 0.8</td>
<td>1.4</td>
</tr>
<tr>
<td>1.3</td>
<td>20.04</td>
<td>2.0</td>
<td>0.8 + 0.8</td>
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<tr>
<td>1.4</td>
<td>19.93</td>
<td>1.8</td>
<td>0.8 + 0.8</td>
<td>0.6</td>
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<tr>
<td>1.5</td>
<td>19.88</td>
<td>1.7</td>
<td>0.8 + 0.8</td>
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To quantitatively determine the effects of compensating GVM$_{si}$ on the gain bandwidth, four different central wavelengths of the seed pulse varying from 1.2 $\mu$m to 1.5 $\mu$m are studied. Figures 2(a)-2(d) present the output spectra of different seeds in both schemes. As the GVM$_{si}$ compensated by BaF$_2$ crystal, all signal bandwidths are extended. For the near-degenerate seed (1.5 $\mu$m), the group velocities of signal and idler are close, thus broadening of the output spectrum is not obvious. But for a seed around 1.3 $\mu$m in non-degenerate range, where the signal and idler wavelengths are distant from each other, large GVM$_{si}$ accumulates during propagation and dramatically narrows the gain bandwidth. By carefully adjusting the length of BaF$_2$ crystal to compensate the GVM$_{si}$ induced in the first amplification stage, gain bandwidth of non-degenerate signals (1.2 $\mu$m~1.4 $\mu$m) is much more effectively enhanced than
that in the near-degeneracy. The compressed temporal intensity profiles of 1.3-μm and 1.5-μm signal pulses are presented in Figs. 2(e) and 2(f), respectively. Pulses centered at 1.3 μm with a sub-three-cycle duration (~10 fs) can be obtained in DOPA scheme, nearly 20% shorter than that in the SOPA, while the 1.5-μm pulse durations are almost the same.

Fig. 2. Output signal spectrum of (a) 1.2 μm, (b) 1.3 μm, (c) 1.4 μm and (d) 1.5 μm seed, and temporal profile of (e) 1.3 μm and (f) 1.5 μm signal pulses after compensation of spectral phase. Blue lines stand for SOPA and red lines stand for DOPA, respectively.

Besides the GVM_{si}, it is also critical to consider the GVM_{ps} and GVM_{pi} for the OPA efficiency. We study the 1.3-μm seed and vary the pump intensity from 40 GW/cm² to 120 GW/cm² for a detailed investigation. The output signal energy and bandwidth for both schemes are compared in Fig. 3. With a low pump intensity, two schemes extract signal pulses with nearly equal energy while the gain bandwidth in DOPA is broader. As the pump intensity increased, the DOPA scheme becomes advantageous with a higher conversion efficiency and a broader signal bandwidth. This phenomenon can be attributed to the gain property of the parametric amplification process. When the pump intensity is low, the amplified pulses are far from saturation and much weaker comparing to the pump, thus the signal energy grows exponentially with the crystal length in accordance with the undepleted-pump approximation. Therefore, with a thicker crystal in the SOPA, the output pulse energy is slightly higher, meanwhile a narrower spectrum is extracted due to the larger GVM_{si} accumulation. For higher pump intensities, the signal gets amplified more efficiently. In SOPA, the signal pulse travels to the weak trailing edge of the pump and approaches saturation rapidly, thus further amplification is restrained. In DOPA, however, the temporal walk-off induced in the first stage is eliminated, so that the amplified pulses can stay under the most intense part of pump, resulting in higher gain in the second stage. Pumped by an intensity of 120 GW/cm², the attained signal energy in DOPA can
reach as much as twice of SOPA according to Fig. 3, corresponding to a conversion efficiency of more than 25%, meanwhile a broad bandwidth supporting sub-three-cycle TL duration is preserved.

Before we continue, we want to evaluate influence of the initial pump-signal delay on the performance of SOPA. The initial pump-signal delay is varied from 5 fs to 40 fs in SOPA, and then the output signal bandwidth and energy are compared to those in DOPA. Since the added delay is beneficial for improving the pump-signal overlap in SOPA, a higher conversion efficiency is achieved. For DOPA, the attainable signal energy can be improve by increasing the thickness of dual-BBO from 0.8 + 0.8 mm to 0.9 + 0.9 mm. Under this condition for achieving high conversion efficiency, the output signal energy and bandwidth in SOPA and DOPA are plotted in Fig. 4.

As shown in Fig. 4(a), even though the conversion efficiency in SOPA is improved by optimizing initial pump-signal delay to 25 fs, DOPA is capable of achieving the same gain with the 1.8-mm BBO, which is still thinner than the 2.0-mm BBO used in SOPA. On the other hand, the signal bandwidth in SOPA is reduced to 185 nm when the optimized pump-signal delay for high energy gain is introduced. Although there is a trade-off between the obtained bandwidth and energy gain when we increase the thickness of dual-BBO, the obtained signal bandwidth
in DOPA is still 25 nm broader than that in SOPA due to the GVM between three interacting pulses are simultaneously compensated. The broader bandwidth in DOPA is resulted from the compensation of not only GVM\(_{ps}\), but also GVM\(_{pi}\) and GVM\(_{ps}\), which cannot be compensated in SOPA by adding an initial pump delay. Therefore, the DOPA scheme is still more favorable for achieving high energy gain and broad bandwidth simultaneously.

4. Superfluorescence and B-integral performance

In the following section, several detrimental effects are investigated to ensure the output pulse quality. During the OPA process, parametric superfluorescence (SF) originated from the amplification of vacuum or quantum noise, also known as optical parametric generation (OPG), must be considered. Despite the availability of generating femtosecond laser pulses, OPG will severely degrade the signal-to-noise ratio (SNR) and CEP stability in an OPA, because of its completely random intensity and phase [40].

![Fig. 5. Energy evolution of 1.3 μm signal in SOPA (blue line) and DOPA (red line), noise energy plotted below (green line) in logarithmic coordinates.](image)

In order to determine the influence of OPG in DOPA, we assume the SF field with an energy of approximately 60 pJ to simulate an initial condition close to the experiment in [41], and the energy evolution of signal and noise in both schemes is evaluated. The result is plotted in Fig. 5. It can be seen from the figure that the noise energy growth follows the same rule in two schemes, which coincides with the undepleted-pump approximation. For the signal pulses, in SOPA scheme, the large temporal walk-off leads to rapid saturation of the amplified pulses, while in DOPA, the compensation of GVM\(_{ps}\) and GVM\(_{pi}\) improves three-pulse overlap, the signal and idler pulses can stay under the central part of the pump and get continuously amplified instead of reaching saturation. As a result, the SNR in our scheme is approximately one order of magnitude higher when the same signal gain is attained. The result can be owed to two crucial factors. First, the SNR is largely dependent on the temporal overlap between the pump, seed, and noise. Since OPG only occurs in the time window defined by the pump, the central part of the SF noise under the pump peak may be amplified more efficiently than the signal pulse overlapped with the trailing edge of pump as in the SOPA. Therefore, improving the temporal overlap is helpful to increase the SNR of the yielded pulses. Second, when the sig-
nal approaches saturation in OPA, the gain efficiency of the signal declines while the relatively weak noise gets amplified preferentially, resulting in rapid decreasing of the SNR [42]. Thus a higher SNR is achieved in our unsaturated DOPA scheme. The signal energy evolution for pump-delayed SOPA is also calculated, even though the signal gain efficiency is improved in SOPA, the SNR in DOPA is still one order of magnitude higher because thinner BBO crystals are employed.

Figure 6 compares the temporal profile of the output signal pulses in two schemes. Since the amplified pulses propagate slower than the pump in BBO crystal, the large temporal walk-off accumulated in the SOPA leads to an intense side lobe (pointed out by the blue arrow) at the leading edge of the signal. In the DOPA, however, two edges of the signal are amplified in each stage respectively (pointed out by the red arrows), although the side lobes are also amplified, the resulted intensity is over two orders lower than that in the SOPA. Besides, as the temporal overlap optimized in our scheme, signal pulse stays localized at the center of the pump, amplifying its peak intensity to a higher level with weaker noise, therefore a pre-pulse contrast around $10^6$ is obtained.

Figure 7. B-integral buildup for 1.3 μm signal pulses in SOPA (blue line) and DOPA (red line).
For interactions between high-intensity pulses in a nonlinear crystal, other undesirable effects caused by Kerr nonlinearity may occur during pulse propagation and amplification. Here, a parameter named B-integral is introduced to evaluate these effects [43]. The B-integral in OPA can be defined as

\[ B = \frac{2\pi}{\lambda_s} \int_0^L n_2(\gamma_s I_s(z) + \gamma_p I_p(z) + \gamma_i I_i(z))dz \]  

where \( \lambda_s \) is the signal wavelength, \( L \) is the nonlinear crystal length, \( n_2 \) is the nonlinear refractive index of the crystal, and \( I_j \) \( (j = p, s, \text{ and } i) \) is the position-dependent intensity of the pump, signal, and idler pulses, respectively. Note that \( \gamma_{mn} (m, n = p, s, \text{ and } i) \) are the correlation factors for a Kerr-effect nonlinearity accounting for self-phase-modulation (SPM, \( m = n \)) and cross-phase-modulation (XPM, \( m \neq n \)). Value of the correlation factor depends on the polarization of two waves: for two waves with the same polarization, \( \gamma_{mn} \) is equal to 1, or else it is 1/3. For a detailed investigation, the value of B-integral is calculated during the numerical simulation process. Simulation parameters are the same that we used in Sec. 3 for the 1.3-\( \mu m \) seed. The calculation result is plotted in Fig. 7. It was pointed out in [43,44] that the B-integral is better to be limited under 1 to ensure a good spatial-temporal quality of the yielded pulses. As can be seen from Fig. 7, B-integral in DOPA is suppressed to 0.86, 30\% lower than 1.22 in SOPA, even if a higher intensity of the signal pulse is obtained. For this reason, better spatial and temporal beam quality can be expected in our scheme. In addition, based on our calculation of the B-integral buildup in pump-delayed SOPA and DOPA, the B-integral has increased to 1.32 and 1.16 in optimized SOPA and DOPA, respectively.

5. Idler pulse analysis

Since the WLC seed inherits the CEP of the pump in OPA, the possibility of generating passively phase-stabilized pulses draws our attention to the idler [45]. As reported in many previous works, difference-frequency idler can be amplified in subsequent OPA stages to obtain ultra-broadband high-intensity CEP-stable pulses [19,20,25,46-50]. Here, we employ a collinear geometry in DOPA to extend the idler gain bandwidth while avoid the angular dispersion of idler pulses.

Based on the energy conservation law, it is reasonable to deduce that the gain bandwidth and conversion efficiency of the idler can be improved similar to the signal by compensating the GVM,\( \gamma_{ij} \). In order to confirm this assumption, the idler pulses are studied by simulating the OPA process using the same parameters in Table 1. It is worth noting that the transparency range of BBO crystal at high-transmittance level (> 90%/2 mm) only covers 0.21 \( \mu m \) to 2.1 \( \mu m \), and the absorption for 2.1 ~ 2.3 \( \mu m \) is crucial. A large portion of >2.1 \( \mu m \) spectral components will be absorbed by the crystal, degrading the gain performances for idler at longer wavelength. Taking the absorption of BBO into account, the output spectra of idler pulses centered at different wavelengths are presented in Figs. 8(a)-8(d), and the compressed temporal intensity profiles of 2.1-\( \mu m \) and 1.7-\( \mu m \) idler are shown in Figs. 8(e) and 8(f). It is observed that broadening of the idler spectra follows the same rule as the signal pulses exhibited in Fig. 2, and the non-degenerate idler at 2.1 \( \mu m \) obtained from DOPA is over 20\% shorter than that in the SOPA, corresponding to sub-two-cycle (~11 fs) pulses. Besides, as shown in Fig. 8(a), spectrum of idler pulses with >2.1 \( \mu m \) components is affected by the BBO absorption, which limits the DOPA tunability at longer wavelengths. Taking the BBO absorption into account, idler pulses centered at 2.1 \( \mu m \) with an energy of 260 \( \mu J \) are extracted, and if the pulses are driven to saturation with thicker dual-BBO, the idler energy will be 565 \( \mu J \). By using higher pump intensity (200 GW/cm\(^2\)) and larger beam size (20 mm diameter), the idler energy can reach as much as 4.8 mJ with a sub-two-cycle TL duration.
Another attractive feature of idler pulses is the passively stabilized CEP [45], which is decisive in many applications. To determine the CEP shot-to-shot variation in DOPA, we simulated the parametric amplification process repeatedly for 500 times, in which the SF field has completely random intensity and phase, and the initial CEP of seed pulse is arbitrary each time. The CEP stability of idler pulses is shown in Fig. 9. Idler with a CEP fluctuation of 97.8 mrad RMS is obtained in DOPA scheme, less than the 125.7 mrad RMS in the SOPA. Due to the better suppressed SF amplification, the CEP of idler pulses produced in our scheme is more stable. As a result, the DOPA turns out to be an ideal source providing few-cycle CEP-stable pulses in the MIR range without any CEP-stabilizing stages.

6. Conclusion

We proposed a collinear OPA scheme employing a compact dual-crystal configuration for GVM-compensating. In this scheme, GVM emerged in the first stage can be effectively compensated by BaF2 crystal, significantly improving the temporal overlap in the second stage. The simulation result shows that ultrashort pulses tunable in the non-degenerate region can be obtained. By carefully adjusting the crystal length and the input pump intensity, our scheme is capable of generating signal pulses centered at 1.3 μm with a broad bandwidth supporting sub-three-cycle duration and a conversion efficiency of over 25%. Besides, CEP-stabilized MIR idler pulses free of angular dispersion are produced, corresponding to sub-two-cycle (∼11 fs at 2.1 μm) TL duration. The detrimental effects originated from parametric SF and Kerr nonlinearity are also studied, higher pulse contrast, better beam quality and less B-integral accumula-
tion can be expected in the DOPA. Besides the BBO crystal, the DOPA scheme is potentially available for other nonlinear crystals, such as lithium triborate (LBO) and lithium niobate (LN). By calculating the dispersion characteristics of these crystals, GVM in LBO and LN crystal can also be compensated by BaF$_2$. Since LBO has the highest damage threshold in all commonly-used nonlinear crystals, it is an ideal candidate for DOPA with higher pump intensity. Because of the wide transparency range of LN crystal, it is a good choice for the generation of IR pulses covering 2 $\mu$m$\sim$4 $\mu$m region. With optimized DOPA, few-cycle signal pulses tunable from 1.2 $\mu$m to 1.6 $\mu$m and idler pulses tunable from 1.6 $\mu$m to 2.1 $\mu$m with millijoule-level energy can be readily achieved, and the extension of this scheme to generate CEP-stable pulses at longer wavelengths can be expected. We believe that the DOPA with designed parameters will not only contribute to the ultrafast coherent x-rays generation but also strong-field laser physics.

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