Nonlinear spectral-phase-engineering strategies via quasiparametric chirped-pulse amplification

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I. INTRODUCTION

Petawatt light (1 PW \(=10^{15}\) W) sources have been applied for high-energy ion generation, laser-driven ion acceleration, and laser weak-field electron acceleration [1–4]. Optical parametric chirped-pulse amplification (OPCPA) is widely used in full-OPCPA PW-class laser facilities or as a front end in a hybrid PW-class laser for its potential in providing high-energy and ultrashort pulses [5–7]. A key task in high-intensity CPA or OPCPA laser is high-order dispersion management [8]. To achieve the Fourier-transform limit (F-T limit) in femtosecond petawatt facilities shorter than 30 fs, the third-order dispersion (TOD), as well as the fourth-order dispersion (FOD) must be well compensated [9]. The main source of TOD and FOD is petawatt laser facilities is from stretchers and compressors, i.e., grating pairs [10]. Several methods have been developed to compensate for the TOD and FOD induced by compressors and material dispersion, such as well-designed tiled gratings [11] or grism pairs [9]. These parts are designed to precompensate high-order dispersions as stretchers in PW-class laser facilities. Both TOD and FOD can be well compensated in an ideal system by configuring the tilt angle and position of gratings, as well as the angle of the incidence beam [12,13]. In a real system, an additional part is often necessary for compensating the residual nonlinear phase, such as an acousto-optic programmable dispersive filter (AOPDF) [14] or grism pairs [13], with intensity limitations and considerable intensity losses.

The nonlinear process in OPCPA has been discussed for over 20 years [15]. With the nonlinear optical parametric amplification (NOPA) technique, few-cycle pulses can be easily generated by the broadband parametric amplification [16]. However, the conversion efficiency of OPCPA is limited to 20% due to the back conversion [17]. To improve conversion efficiency, a new method is developed by absorbing idler to suppress the back conversion called a quasiparametric chirped-pulse amplifier (QPCPA) [18–21]. Using this technique, the energy conversion efficiency can be performed up to 41%. Recent results show the potential of QPCPA in high efficiency and high stability amplification [19]. Meanwhile, to the best of our knowledge, no one considered the significant difference between the nonlinear phase induced by QPCPA and the nonlinear phase induced by OPCPA.

Here in this article, we will show a method for nonlinear spectral phase compensation via QPCPA. The high-order dispersions can be well compensated by adjusting the phase mismatch, by applying a QPCPA preamplifier module. In the following sections, we will discuss the theoretical models, giving a concise expression to the absorption-induced optical parametric phase under different conditions. At the same time, we will analyze the compensation process with the help of numerical simulation and discuss this method affecting pulse shaping in the parametric amplification process.

II. THEORETICAL ANALYSIS

The three-wave mixing process of OPCPA or QPCPA can be described by a set of coupled-wave equations which can be derived under the slowly varying envelope approximation from [22]:

\[
\frac{\partial A_i}{\partial z} + \sum_{n=2}^{\infty} \frac{(-j)^{n-1}}{n!} \frac{\partial^n k_i}{\partial \omega^n} \frac{\partial^n A_i}{\partial \tau^n} + F^{-1}\left(\frac{1}{2} \alpha_i(\omega)\right) * A_i = j 2d_{eff} \omega_0^2 \frac{\partial^2 A_i A_p}{\partial \tau^2} + F^{-1}\left(\frac{1}{2} \alpha_i(\omega)\right) * A_i
\]

\[
\frac{\partial A_i}{\partial z} + \frac{\partial k_i}{\partial \omega} \frac{\partial A_i}{\partial \omega} + \sum_{n=2}^{\infty} \frac{(-j)^{n-1}}{n!} \frac{\partial^n k_i}{\partial \omega^n} \frac{\partial^n A_i}{\partial \tau^n} + F^{-1}\left(\frac{1}{2} \alpha_i(\omega)\right) * A_i = j 2d_{eff} \omega_0^2 \frac{\partial^2 A_i A_p}{\partial \tau^2} + F^{-1}\left(\frac{1}{2} \alpha_i(\omega)\right) * A_i
\]
where \( A_m (m = s, i, p) \) are the complex pulse envelopes of the signal, the idler, and the pump light, respectively. \( \tau \) is the normalized time with the pulse duration of signal \( t_s \). \( \Delta k_0 = k_s + k_i - k_p \) is the phase mismatch, \( d_{\text{eff}} = \chi_{\text{eff}} (2) / 2 \) is the effective nonlinear coefficient, \( c \) is the speed of light, and \( \omega_{s,i,p} \) are the angular central frequency of the signal, the idler, and the pump, respectively. \( \alpha_{s,i,p} (\omega) \) are the absorption coefficients. For OPCPA setup, \( \alpha_{s,i,p} (\omega) = 0 \), and in QPCPA, the absorption of idler \( \alpha_i (\omega) \neq 0 \). \( F^{-1} (1 / 2 \alpha_{s,i,p} (\omega)) \) are the inverse Fourier transform in the temporal domain of the absorption coefficient of the signal, the idler, and the pump, respectively. \( F^{-1} (1 / 2 \alpha_{s,i,p} (\omega)) \) is the convolution of absorption and the complex pulse envelopes in the time domain.

Considering a noncollinear setup as shown in Fig. 1 with a small-signal approximation and ignoring the pump light depletion, the intensity-induced nonlinear spectral phase of the signal can be analytically expressed by Eq. (4), which is known as the optical parametric phase (OPP) [23]:

\[
\varphi_{\text{OPP}} (\omega_s) \approx - \arctan \left( \frac{\Delta k'}{\Gamma_{\text{OPP}}} \right),
\]

when \( \Delta k_0 < 2G \),

\[
\varphi_{\text{OPP}} (\omega_s) \approx - \arctan \left( \frac{\Delta k'}{\Gamma_{\text{OPP}}} \right),
\]

when \( \Delta k_0 > 2G \).

Here \( \Gamma_{\text{OPP}} = \sqrt{G^2 - \Delta k_0^2 / 4} \) and \( h = \sqrt{-G^2 + \Delta k_0^2 / 4} \), where \( G \) is the gain factor. This approximation can hold when the depletion of pump intensity is less than 10% in our calculation.

Using the same presumption with Eq. (4) when \( \Delta k_0 < 2G \) and including the absorption of the idler and the group delay of signal and idler, the absorption-induced OPP (AIOPP) can be expressed as

\[
\varphi_{\text{AIOPP}} = - \arctan \left( \frac{\Delta k'}{\Gamma_{\text{AIOPP}}} \right) - g \zeta \quad \text{when } f \gg g \quad \text{and} \quad e^{-2\zeta f} \ll 1,
\]

where the parameter \( C_1 = \alpha_s g / 2 \Delta k' f, C_2 = 2f^2 + \alpha_s f / 2 - g (\Delta k' - 2g), C_3 = [(2f + \alpha_s / 2)^2 + (2g - \Delta k')^2]G^{-2}e^{-2\zeta f}, \) the real part \( f = \text{Re}(\sqrt{G^2 - (\Delta k')^2 / 4} + (\alpha_s / 4) f) \), and \( g \) is the corresponding imaginary part. Here we define \( \Delta k' - \Delta k_0 = k_i (\Delta \omega + \alpha_i) + k_i (\Delta \omega - \alpha_i) - k_p (\omega_p) - \Delta k_0 = \sum_{n=1}^{\infty} \left( \frac{\alpha_n}{\Gamma_{\text{AIOPP}}} \right)^n (\Delta \omega)^n \), \( \Delta \omega = \omega - \omega_s \). Notice that the term \( \sqrt{G^2 - (\Delta k')^2 / 4} + (\alpha_s / 4) f \) is always complex when \( \alpha \neq 0 \). Without absorption, this term will be either a real number (when \( \Delta k < 2G \)) or an imaginary number (when \( \Delta k > 2G \)), and the AIOPP becomes OPP in this case. The influence of pump power to AIOPP is complex, which makes the AIOPP an extraordinarily complex expression. When the induced phase is much smaller than the gain factor in quantity, and \( e^{-2\zeta f} \ll 1 \), the terms \( C_1 \) and \( C_3 \) will be much smaller than \( C_2 \) in Eq. (5), and the AIOPP can be approximated to \( -g \zeta \). In order to compare the difference between the spectral phase in OPCPA and QPCPA, the same term \( -\alpha_i \Delta k'/2G_2 \) is excluded. This term only contains dispersions up to the third order in the discussion and thus can be excluded when focusing on higher-order dispersions.

III. RESULTS AND DISCUSSION

First we discuss the case of QPCPA working with near-perfect phase matching when \( \Delta k' \ll 2\sqrt{G^2 + \alpha_i^2 / 16} \) and \( Gz > 1 \). In this case, the real part \( f \approx G' \) and the imaginary component \( g \) is simplified to the expression \( -\alpha_i \Delta k'/8G' \). Here \( G' (\omega) = \sqrt{G^2 - \Delta k_0^2 / 4 + \alpha_i^2 / 16} \) and \( G' = G' (\omega_i) \).

To qualitatively investigate the nonlinear spectral phase, we rewrite the AIOPP with Taylor expansion as

\[
\varphi_{\text{AIOPP}} = \varphi_{(0) \text{AIOPP}} + \sum_{n=1}^{4} \frac{\varphi_{(n) \text{AIOPP}}}{n!} (\Delta \omega)^n + o(\Delta \omega^5).
\]

Consider an absorption spectrum with a Gaussian shape,

\[
\alpha_i = \alpha_{i0} \exp(- (\omega - \omega_i)^2 R^2).
\]

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The even-order dispersions and odd-order dispersions are largely affected by the phase mismatch $\Delta k_0$ and the group velocity mismatch (GVM) $\beta'_0$ in QPCPA, respectively.

When a final stage amplifier uses QPCPA, where the pump is rapidly depleted and the idler absorption will cause a large temperature rise of about 100 K, the temperature-induced phase mismatch can largely affect the high-order dispersions. The following approximations:

$$\phi^{(2)}_{\text{AIOPP}} \approx -2! \frac{\alpha_0 \beta'_0}{G} \frac{G^2 - \alpha_0^2}{8} \frac{\Delta k_0}{z^2},$$

$$\phi^{(3)}_{\text{AIOPP}} \approx -3! \frac{\alpha_0^2 \beta'_0^2}{G^3} \frac{G^2 - \alpha_0^2}{8} \frac{\Delta k_0}{z^3}$$

$$\phi^{(4)}_{\text{AIOPP}} \approx 4! \frac{\alpha_0^3 \beta'_0^3}{G^4} \frac{G^2 - \alpha_0^2}{16} \frac{\Delta k_0}{z^4}$$

When the group velocity of signal and idler are matched, the residual spectral phase $\phi(\omega)$ can be ignored, the expression can be used to solve the proper residual-phase-eliminated conditions:

$$\phi(\omega) = \frac{\alpha_0 \Delta k_0}{8 G} \left( \frac{G^2 - \alpha_0^2}{8} \frac{\Delta k_0}{G^3} + \frac{(G^2 - (\alpha_0/4)^2)^2}{8 G^3} \right) \frac{\Delta k_0}{z^2}$$

Residual spectral phase contributions should not have a large impact within the whole spectrum range to reach the F-T
limit. The exact number of FOD can only be calculated with the accurate expression in Eq. (5) when both $\alpha_0$ and $\Delta k_0$ are comparable with $G$, even though Eq. (13) has enough accuracy to estimate the residual-phase-eliminated conditions.

Considering there is only group delay $\beta'_1$ in the system to compensate the FiOD in the signal pulses, the high-order dispersion-eliminated conditions are almost the same with FOD compensation. The linear relation of $G$ and $\alpha_0$ is determined by the ratio $K$ [Fig. 3(a)]. When the gain factor is nearly flat within the whole spectrum, the FOD control range is limited by the bandwidth of the absorption spectrum, showing the requirement of large-absorption-bandwidth nonlinear crystals. Recently, Yang et al. has reported the preparation process of Pr : LiNbO$_3$ crystals with broadband absorption [24]. The ratio $K$ depends on the spectrum range where the residual phase should be controlled, and a larger spectrum range requires a large absorption. On the other hand, the condition of eliminating $\alpha(\omega^2)$ is less affected by $\Delta k_0$, $\beta'_1$, and $z$, which ensures the feasibility of engineering the FOD and FiOD in a QPCPA preamplifier.

A NOPA geometry can offer a large gain bandwidth when the angle a between the signal beam and the pump beam has the following relationship: $\alpha = \sin^{-1}[(1 - v_s^2/v_i^2)/(1 + 2v_s k_s/v_i k_i + k_s^2/k_i^2)])^{1/2}$ [25], where $v_s$ and $v_i$ are the group velocity of signal and idler, respectively. FOD control by QPCPA requires the same phase mismatch within the spectrum so that NOPA is the best geometry compared to other geometries, such as quasi-phase matching or noncritical phase matching. By adjusting the crystal rotation to the desired angle in a NOPA geometry, it is able to adjust the FOD between $-3 \times 10^5$ and $3 \times 10^5$ fs$^2$ and FiOD between $-3 \times 10^7$ and $3 \times 10^7$ fs$^2$ in a 10-mm length nonlinear crystal (for example, Sm:YCOB) for ~40-fs signal pulses. When adjusting the FOD between $-3 \times 10^5$ and $3 \times 10^5$ fs$^2$, the corresponding group delay dispersion (GDD) changes between $-500$ and $500$ fs$^2$. This GDD change can be easily compensated by adjusting grating pairs or prism pairs with only negligible effect on FOD, and the compensator can be designed as an adjustable device. Moreover, the beam direction will not change after the compensation of FOD. FiOD can be controlled with $\beta'_1$ in the same way but will also cause gain narrowing when $\beta'_1$ is too large.

To explore the properties of the QPCPA process in the temporal region, numerical simulations are done with Eqs. (1)–(3) based on the split-step Fourier-transform method combined with the Runge-Kutta method. This kind of modified split-step-Fourier method has been proved to be more accurate than the merely split-step-Fourier method [26].

Results show that the signal spectrum is broadened because the QPCPA gain factor is higher in the sides of the spectrum rather than higher in the center, and the gain factor is not largely affected by the phase mismatch. The QPCPA outputs shorter pulses than the original pulses after the compressor, as is shown in Fig. 4(a). Presuming a laser system with FODs of $3 \times 10^5$ fs$^4$ in 40-fs signal pulses, a QPCPA preamplifier can replace the OPCPA preamplifier to compensate FOD with $G = 320$ m$^{-1}$ and $\Delta k_0 \approx 300$ rad/m in 10-mm crystals. Figure 3(b) shows the residual phase can be well compensated in the whole spectrum range. To show the limit of the FOD control ability, a QPCPA with a gain factor $G = 640$ m$^{-1}$, $\Delta k_0 \approx 1000$ rad/m, and $\alpha_0 = 20$ cm$^{-1}$ is also calculated. In this parameter, a FOD up to $1.2 \times 10^9$ fs$^3$ can be well compensated. Although these FODs can be compensated by adjusting GDD in a certain degree as well, FODs can be better controlled with QPCPA phase modulation. The residual phase controlled by QPCPA is flatter than merely adjusting the GDD, which results in shorter pulses with a better pulse contrast (an order higher at $\pm 400$-fs scale). The AIOPP in a QPCPA preamplifier is not affected by the intensity of signal pulses when its intensity is in the small-signal region so that the AIOPP will not be affected by spatial beam profiles or intensity fluctuations of the signal.

Although the ideal phase compensation parameters we get from the above analytical expressions are a specific point, numerical results show the best phase compensation parameters can cover a wide range. We have calculated the pulse width of QPCPA output ($t_{QPCPA}$) and the corresponding Fourier-transform limits ($t_{F-T}$) among various conditions. $\Delta t = t_{QPCPA} - t_{F-T}$ can be used to characterize the phase compensation in QPCPA. The ideal parameters in Fig. 4(a) are $G = 320$ m$^{-1}$ and $\Delta k_0 = 300$ rad/m, used to compensate for an initial FOD of $3 \times 10^5$ fs$^2$, and both the gain factor and phase mismatch can be adjusted in a wide range without

FIG. 3. (a) Absorption requirement for eliminating high-order dispersions within the range of $\pm 1.5\Delta k_0$. (b) Spectral phase of QPCPA with large phase mismatch. The curve is calculated with $z = 6$ mm, $G = 400$ m$^{-1}$, $\alpha_0 = 7.46$ cm$^{-1}$, and $\Delta k_0 = 200$ rad/m, with the absorption spectrum bandwidth equal to the bandwidth of the signal. The signal has a central wavelength of 810 nm, and the Fourier limit is 40 fs. The pump intensity is about 4 GW/cm$^2$ with 532 nm.

$$G \approx K\alpha_0$$

$$\Delta k = 200, 100, 50$$

Absorption (arb. units)

Gain parameter G

Normalized Intensity (arb. units)

$\alpha_0 = 5$ 

$G = 640$ m$^{-1}$ 

$\Delta k_0 \approx 1000$ rad/m 

$\alpha_0 = 20$ cm$^{-1}$

$\beta'_1$

$z$
large effects on the pulse duration. The widest adjustable range can be achieved in Fig. 4(a) when $\Delta k_0 \approx 300 \text{rad/m}$ and $G \approx 250 \text{m}^{-1}$. Although the FODs provided by QPCPA in these regions is far from the initial FOD, the residual phase after GDD compensation is still well controlled, which shows the high adaptability of this FOD compensation scheme. The thermal effect of idler absorption is less than 0.1 K in our calculation at a repetition rate of 100 Hz (calculated by the same method in Ref. [18] when the output signal is $\sim 70 \text{mJ}$ and beam radius is 2.5 mm with an idler absorption of about 8 cm$^{-1}$), and the phase mismatch caused by idler absorption can be ignored ($\Delta k_0$ changes $<1 \text{m}^{-1}$). Figure 5(b) shows the changes of $\Delta t$ with the phase mismatch and the initial FOD in the signal pulses when the gain factor $G$ is fixed at 320 m$^{-1}$. The initial FOD between 0 and $3 \times 10^5 \text{fs}^4$ can be well compensated by varying the phase mismatch $\Delta k_0$.

A set of two-dimensional simulation results is shown in Fig. 6 to discuss the system stability towards pump power fluctuations or the pump intensity distributions. The pump beams in recent OPCPA have high-intensity uniformity with top-hat intensity profiles. QPCPA enlarges the high-order dispersions by idler absorption, and the AIOPP is sensitive towards the pump intensity, so QPCPA also requires a top-hat intensity profile pump laser. The QPCPA phase compensation process shows high stability towards the gain factors changes. To clarify this point, we show the comparison between the pulse duration with FOD compensated by QPCPA and the shortest pulse duration with FOD compensated by excessive GDD. Results show that the pulse duration can be kept to the F-T limit, even when the pump intensity decreases to 65% of the maximum. Spectrum modulation also plays an important role in the compression process. The intensity is concentrated in the two sides of the QPCPA spectrum, which is more insensitive towards even-order dispersions compared with a Gaussian-like spectrum. While pulse duration which is compressed by only excessive GDDs is sensitive towards spectrum changes, the pulses compressed by QPCPA are insensitive towards spectrum changes and can keep the pulses near the F-T limit when the pump intensity or signal intensity changes.

Although the compensating process is limited by absorption bandwidth, it is possible to compress a short pulse with narrower absorption bandwidth. We use both the difference between the pulse duration of QPCPA outputs and its F-T limits ($t_{\text{QPCPA}} - t_{\text{F-T}}$) and the difference between the pulse duration of QPCPA outputs and the F-T limits of original signals $t_{\text{QPCPA}} - t_{\text{signal}}$ to characterize the pulse compression performances. Results show that a signal pulse of 20 fs can be well compressed by QPCPA with an 89-nm bandwidth absorption spectrum ($\omega_s/\omega_{ai} = 2$) and $1 \times 10^5 \text{fs}^4$ FOD [Fig. 7(a)]. If the 20-fs signal carries a large FOD (for example, $3 \times 10^5 \text{fs}^4$), the QPCPA cannot compress the pulse into...
FIG. 6. (a)–(c) Intensity profile of the signal, a top-hat pump, and a top-hat pump with a larger beam radius, respectively. (d) Difference between the compressed pulse duration and the corresponding F-T limits. The compression GDD is fixed at $-652.66 \text{ fs}^2$ in both QPCPA and excessive GDD phase compensation. The curves are for the QPCPA process (green) and the AIOPP added on the input Gaussian-like spectrum (blue); the FOD compressed by the GDD with a spectrum of QPCPA output (orange) and input (brown); the OPCPA process with excessive GDD compression (wine), respectively. (e), (f) Compressed pulse duration calculated with a top-hat pump covering the center of the signal and the compressed pulse duration calculated with a top-hat pump covering nearly the whole signal, respectively. (e), (f) Calculated with $G = 450 \text{ m}^{-1}$ and $\Delta k_0 = 180 \text{ rad/m}$, and the pulse duration of the initial signal is 20 fs.

QPCPA F-T limits. But it is still able to reach the F-T limit of the original signal. To show the limitation of the FOD control of short pulses, a broadband absorption ($\Delta \omega_0 / \Delta \omega_{ai} = 1$) with $\alpha_0 = 20 \text{ cm}^{-1}$ is applied to compress $7.5 \times 10^4 \text{ fs}^4$ of 20-fs pulses, which has an F-T limit of 15 fs after being QPCPA amplified [Fig. 7(b)]. The initial FOD with the QPCPA spectrum cannot be well compensated with merely excessive GDD without AIOPP because the spectrum is no longer a Gaussian shape and the residual phase is sensitive to this spectrum modulation. The initial FOD can be well compensated by adjusting the GDD with the help of AIOPP, and the pulse duration is able to reach the F-T limit. However, this FOD control method cannot support a pulse duration shorter than 15 fs in our calculation, because this method requires the same...

FIG. 7. (a) The normalized signal pulse intensities and corresponding phase intemporal region. The GDD and TOD of the QPCPA output are compensated. The initial FOD is $1 \times 10^5 \text{ fs}^4$, gain factor $G = 320 \text{ m}^{-1}$, and $\Delta k_0 = 200 \text{ rad/m}$, $\alpha_0 = 7.67 \text{ cm}^{-1}$, $z = 10 \text{ mm}$. (b) Pulse duration calculated with an initial FOD $7.5 \times 10^4 \text{ fs}^4$ with a 20-fs seed. $G = 640 \text{ m}^{-1}$, $\Delta k_0 = 1000 \text{ rad/m}$, $\alpha_0 = 20 \text{ cm}^{-1}$, $z = 10 \text{ mm}$. The F-T limit (blue) of QPCPA output is 15 fs. A well-compressed pulse (yellow) can be formed from QPCPA output, with GDD and TOD compensated. The pulse cannot be well compensated if considering the initial FOD compressed by merely GDD with such a spectrum without AIOPP.
phase mismatch within the whole spectrum, but a broader spectrum will lead to a rapid change of the phase mismatch, especially when $\Delta k_0$ should be kept large. Moreover, the absorption bandwidth would also be a problem. Even though an absorption spectrum formed by multipeaks may still be acceptable and the phase mismatch can be tuned by angular dispersions of signal, the design and adjusting will be complex and challenging. In the controllable region, this method has the potential to be a powerful tool in OPCPA design and adjusting.

### IV. CONCLUSION

In conclusion, we have shown that the quasiparametric amplification process will lead to strong high-order dispersions. In the small-signal region, the residual phase amplification process will lead to strong high-order dispersions. The FOD can be eliminated when the FOD is controlled, showing the great potential of the QPCPA preamplifier as an efficient amplifier. In the small-signal region, the residual phase amplification process will lead to strong high-order dispersions.

$\sim -3 \times 10^5 \text{ fs}^4$ for $\sim 40$-fs pulses and $\sim 1 \times 10^5 \text{ fs}^4$ for pulses shorter than 20 fs by adjusting the phase mismatch in a 10-mm-long nonlinear crystal. This phase compensation strategy shows a highly efficient, economic, and easy way of operation towards high-order dispersion management in ultrafast laser facilities. Numerical simulation results show that the signal spectrum is broadened because of the phase mismatch insensitive gain factor and thus could get a shorter pulse after the compressor. The compensator is very stable, which allows for wide-range changes in both phase mismatch and the pump intensity. Finally, we point out that the best bandwidth of the absorption spectrum is the same as the idler spectrum, and signal pulses longer than 15 fs with large FODs can be well compressed with the dispersion control.

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