

Communication

# Suppression of Pulse Intensity Dependent Dispersion during Nonlinear Spectral Broadening with Intermediate Compression for Passive CEP Stable Pulse Generation

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**Abstract:** The intensity fluctuation induced spectral phase-change of the laser pulse during nonlinear spectral broadening is theoretically investigated. The oscillation of the phase-change curves at the central part of the spectra is explained by the two-wave interference model, while the bending of the phase-change curves at the wings is considered to originate from the intensity dependent dispersion caused by the self-steepening (SST) effect. Both of them can degrade carrier envelop phase (CEP) stability after an intra-pulse difference frequency generation (IP-DFG) setup. We propose an effective approach to suppress the intensity dependent dispersion with intermediate compression. Verified by numerically simulations, well-phased spectral components at the wings can be obtained, which is highly beneficial for CEP stable pulse generation with noisy input.

**Keywords:** nonlinear spectral broadening; intermediate compression; intensity dependent dispersion; self-steepening



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## 1. Introduction

The control of electric field of few-cycle pulses is essential for field dependent applications, e.g., strong field physics and attosecond science [1–6]. The electric field control requires light pulses with stable CEP, which can be achieved by CEP stabilization techniques, including active and passive ones. The passive stabilization techniques are mainly based on the combination of nonlinear spectral broadening and DFG [7]. Compared to the active ones, the passive stabilization techniques are fully-optical methods with much simpler configurations. The nonlinear spectral broadening can be realized by schemes based on bulk materials [8], gas-filled hollow core fibers [9], multiple thin plates [10–12], and multi-pass cells [13,14]. The latter three schemes are energy scalable and can support millijoule level pulses. With such pulse energy, the spectral broadened pulses can drive IP-DFG setup, whose configuration is simpler compared to the inter-pulse DFG, and can potentially generate pulses with more stable CEP due to common path for all spectral components [12].

The nonlinear spectral broadening process can be interpreted as four-wave mixing, in which the newly generated spectral components acquire intensity dependent phase-shift (i.e., intensity-phase coupling) in addition to the original CEP. Hence, CEP stable pulse can be achieved after the original CEP is cancelled out by the following DFG procedure assuming the intensity noise is negligible [15]. Unfortunately, the intensity noise inherently exists for all the lasers, and intensity-phase coupling introduces different phase-change to each spectral component during the nonlinear spectral broadening process, leading to an intensity dependent dispersion, which degrades the CEP stability in the passive CEP stable setup. G. Steinmeyer et al. have theoretically and experimentally investigated the

phase-change stability between the interested wavelength ( $f$ ) and its second harmonic ( $2f$ ), defined as intra-pulse coherence [16]. This study shows that the intensity noise leads to severe loss of intra-pulse coherence and can greatly lower the reliability of the active CEP stabilization techniques in case that the CEP is measured based on conventional  $f$ - $2f$  interferometers. Furthermore, in view of passive CEP stable pulse generation, the intra-pulse coherence defined in [16] is not straightforward to predict the CEP stability of idler pulse after IP-DFG, where the intensity-dependent phase-change needs to be examined for all the spectral components, covering the spectral region of both the pump and signal.

In this work, we present numerical study on the intensity dependent phase-change during nonlinear spectral broadening process. We consider a typical case for 30 fs input pulse with spectrum centered at 800 nm from Ti:sapphire laser systems. Such lasers are frequently used to drive CEP stable light source based on IP-DFG [9,17]. Through the analysis of the modulation of phase-change curves, we find that the phase-change curves at the wings of the spectra are smoother, but usually bended by the intensity dependent dispersion caused by SST. We suggest to suppress the SST for obtaining well-phased spectral components at the wings by intermediate compression. The achieved spectral components at the wings with suppressed intensity dependent dispersion are extremely suitable to serve as input for IP-DFG to generate ultra-stable CEP pulses.

## 2. Theoretical Methods

In a conventional IP-DFG system, the spectra of pump and signal pulses are selected by bandpass filters, meanwhile the temporal overlapping is realized by compression with chirped mirrors [9,17]. The pump and signal pulses can be written as

$$\begin{aligned} E_s &= A_s(t + \tau_s)e^{i(\omega_s(t+\tau_s)+\phi_s)} \\ E_p &= A_p(t + \tau_p)e^{i(\omega_p(t+\tau_p)+\phi_p)} \end{aligned} \tag{1}$$

where  $\tau_s$  ( $\tau_p$ ) and  $\phi_s$  ( $\phi_p$ ) are the group delay and CEP of the signal (pump) respectively. The idler pulse can be calculated as

$$\begin{aligned} E_i &\propto A_p(t + \tau_p)A_s(t + \tau_s)e^{i(\omega_p(t+\tau_p)-\omega_s(t+\tau_s)+(\phi_p-\phi_s)-\pi/2)} \\ &\propto A_p(t + \tau_p)A_s(t + \tau_s)e^{i((\omega_p-\omega_s)t+(\omega_p\tau_p-\omega_s\tau_s)+(\phi_p-\phi_s)-\pi/2)} \\ &\propto A_p(T)A_s(T + \Delta\tau)e^{i(\omega_iT-\omega_s\Delta\tau+(\phi_p-\phi_s)-\pi/2)} \end{aligned} \tag{2}$$

where  $\omega_i = \omega_p - \omega_s$ ,  $\Delta\tau = \tau_s - \tau_p$ ,  $T = t + \tau_p$ . Considering the intensity dependent dispersion within the spectral broadening procedure, the idler pulse is now written as

$$\begin{aligned} E_i &\propto A_p(T)A_s(T + \Delta\tau)e^{i(\omega_iT-\omega_s\Delta\tau+(\phi_p-\phi_s)-\pi/2)} \\ &\propto A_p(T)A_s(T + \Delta\tau + (\Delta\tau_s - \Delta\tau_p)) e^{i(\omega_iT-\omega_s(\Delta\tau+(\Delta\tau_s-\Delta\tau_p))+(\phi_p-\phi_s+(\Delta\phi_p-\Delta\phi_s))-\pi/2)} \end{aligned} \tag{3}$$

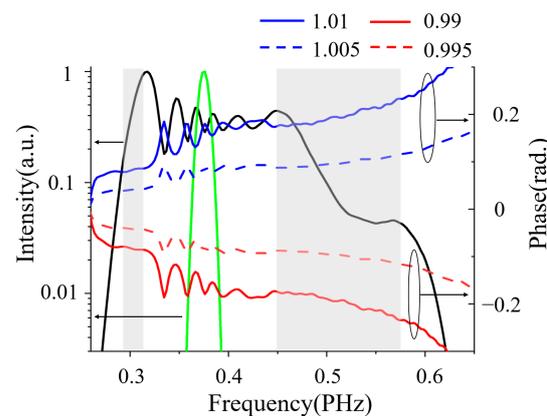
where  $\Delta\tau_s$  ( $\Delta\tau_p$ ) and  $\Delta\phi_s$  ( $\Delta\phi_p$ ) are the variations of group delay and CEP of the signal (pump) caused by the input intensity noise. According to Equation (3), the CEP can be stabilized if the intensity dependent phase-change is linear for frequency in the spectral range of the interacted pump and signal pulse in the DFG. In other words, the intensity noise introduces same group delay and CEP to the pump and signal, thus the intensity dependent dispersion vanishes.

Here nonlinear spectral broadening scheme based on multiple thin plates is considered because of the simpler structure and energy scalability for pulse up to millijoule level. According to [18], the multiple thin plates can introduce quasi-waveguide effect to the pulse, leading to spatially homogeneous spectral broadening. In addition, it has been proved that 1D model is enough to study the physics associated with pulse evolution inside the quasi-wave guide system in a wide range of configurations and nonlinearity levels without knowing the exact configurations [19]. Meanwhile, experimental studies show that spectral broadening with short pulse duration is mainly contributed by the self-phase modulation (SPM) and SST [20]. Therefore, the space-time coupling within the spectral

broadening process is neglected and a 1D model based on carrier-resolved unidirectional optical propagation equation is used to simulate the spectral broadening procedure, in which the dispersion, self-phase modulation and SST are taken into account [21]. In the simulations, transform-limited pulses are taken as input for the nonlinear spectral broadening setup. The nonlinear media employed here is fused silica and peak intensity of  $7.5 \text{ TW/cm}^2$  is used to avoid degradation of the materials. Because the ultra-thin materials are used, self-focusing inside the materials is avoided.

### 3. Results

Firstly, the spectral broadening is realized with single stage setup. The intensity noise is set to 1% in the calculations, which is normal for most of the commercial lasers. The total thickness of fused silica is 0.8 mm. The nonlinear phase of the output pulse is compensated with GDD of  $-40 \text{ fs}^2$ . The difference of the broadened spectra with varied intensity is negligible. Figure 1 shows the input and broadened spectra. The spectral phase-change curves, representing the difference of the spectral phase of different input intensities compared to the one with averaged input, are also showed in Figure 1. The phase-change curves are highly modulated where the spectral intensity oscillates (multiple-peaks structure in Figure 1), which indicates the intensity noise introduces additional high order dispersion to this part of the spectrum. In contrast, the phase-change curves are much smoother beyond the outmost peaks. It is also observed that the phase varies monotonically according to the input pulse intensity, hence it is preferred to have driving laser of ultra-low intensity noise for passive CEP stabilization setup.



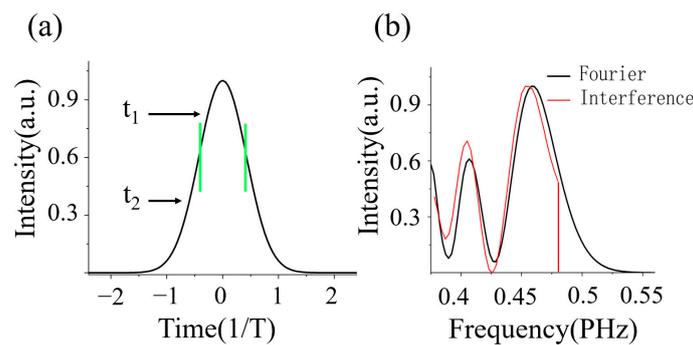
**Figure 1.** Input (green solid line), broadened (black solid line) spectra, and phase-change curves with different input intensity (red line— $0.99 I_a$ , red dash line— $0.995 I_a$ , blue line— $1.01 I_a$ , blue dash line— $1.005 I_a$ , where  $I_a$  is the average intensity). In the shaded rectangles, the phase change curves are close to linear.

In order to capture the origin of the oscillation of the phase-change curves, we investigate the spectral broadening process with a two-wave interference model with only SPM included [22]. With this model, the typical oscillatory character of the broadened spectra and phase-change curves after SPM effect can be analytically analyzed. Figure 2a shows the envelope of transform-limited input pulse with Gaussian temporal shape. The temporally varying nonlinear phase results into a time dependent instantaneous frequency. The spectrum calculated by the interference model is showed by the red line in the Figure 2b. Only half of the spectrum needs to be calculated because of the symmetric spectrum with only SPM considered. The maximum instantaneous frequency shift ( $\nu_{max}$ ) according to the interference model is located at the time with biggest gradient (marked by the green lines in Figure 2a). The gradients at  $t_1$  and  $t_2$  are identical, hence the instantaneous frequencies at these two times both contribute to this spectral component interferometrically in the spectral domain, which causes the oscillation in the central part of the spectrum. Compared to the spectrum calculated by Fourier transform of the electrical field after acquiring the

SPM induced nonlinear phase shift (black line in Figure 2b), one can see that no interference occurs for spectral components beyond  $\nu_0 \pm \nu_{max}$  ( $\nu_0$ —central frequency of the input laser). To analyze the phase modulation, the phase of the spectral component created at  $t_1$  and  $t_2$  can be approximated by

$$\varphi(\omega(t_1, t_2)) = \text{atan}\left(\frac{\sin(\Delta\phi)\sqrt{I(t_2)}}{\sqrt{I(t_1)} + \cos(\Delta\phi)\sqrt{I(t_2)}}\right) \tag{4}$$

where  $\Delta\phi$  is the total phase difference between  $t_1$  and  $t_2$ ,  $I(t_1)$  and  $I(t_2)$  are the intensity at  $t_1$  and  $t_2$ , respectively. The second order derivative of the phase to  $\Delta\phi$  is proportional to  $\sin(\Delta\phi)$ , which means the phase-change reaches the extremum when  $\Delta\phi=0$  or  $\Delta\phi=-\pi$ . From Figure 1, it is clear that the phase-change reaches the peak (marked by green solid lines) when destructive interference happens ( $\Delta\phi=-\pi$ ), while it reaches the valley when constructive interference happens ( $\Delta\phi=0$ ). Since no interference occurs at the wings, the phase-change oscillation is absent. However, SST introduces intensity dependent refractive index term to the pulse [23], which imposes intensity dependent dispersion. When SST is considered, the phase-change curves are bended (Figure 1). With noisy driving lasers, this intensity dependent dispersion will greatly degrade the CEP stability according to Equation (3). This point of view coincides with the statement in [16] that the asymmetric spectral broadening needs to be avoided to mitigate the loss of intra-pulse coherence, while it is a common knowledge that the asymmetric spectral broadening originates from SST.



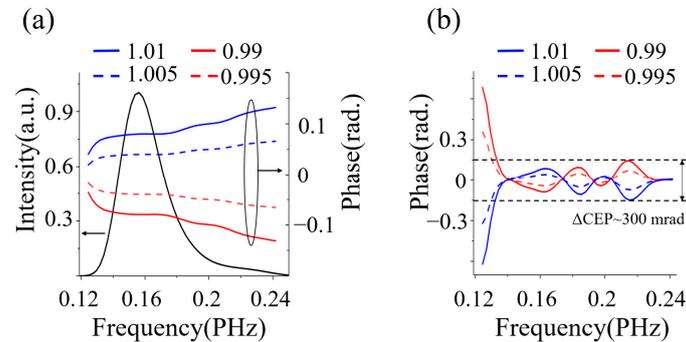
**Figure 2.** The temporal profile of a gaussian pulse (a), the spectral shapes calculated by two-wave interference model and Fourier transform (b).

To evaluate the influence of the intensity noise induced dispersion on the idler CEP after the DFG, the idler pulse is obtained in the un-depleted regime through 1D calculations of the coupled wave equations at perfect phase-matching condition [24]. We assume perfect phase-matching in the calculations to exclude the impacts of the DFG process. The spectral components of the interaction are showed in the grey area in Figure 1, which are chosen in accordance with the experimental studies [9,17]. The idler spectrum and phase-change curves are presented in Figure 3a. The spectrum spans from 0.12 PHz to 0.24 PHz. It is sufficient to support sub-two cycle pulses. To analyze the CEP fluctuation, the frequency resolved CEP changes with different intensity noise level are calculated by

$$\Delta\text{CEP}(\omega) = \Delta\varphi(\omega) - \omega\Delta\tau(\omega) \tag{5}$$

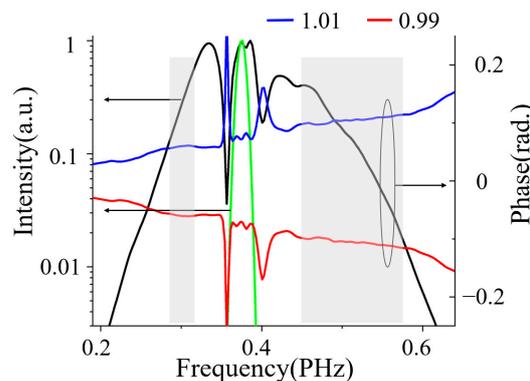
where the  $\Delta\varphi(\omega)$  is the phase-change, and the frequency dependent group delay is  $\Delta\tau(\omega) = \partial\Delta\varphi(\omega)/\partial\omega$ . It can be seen that the CEP fluctuation increases gradually with the increase of intensity noise level. The CEP fluctuation is not uniform for all the spectrum components and the biggest CEP fluctuation range reaches ~300 mrad for spectral components from 0.14 PHz to 0.24 PHz. In addition, the CEP fluctuation increases dramatically towards the red side (more than 1.2 rad at 0.12 PHz). In case the full band is used to gen-

erate sub-two cycle pulses, the CEP fluctuation leap at the red side can lead to significant measurement error of CEP when the f-2f interferometers are used.



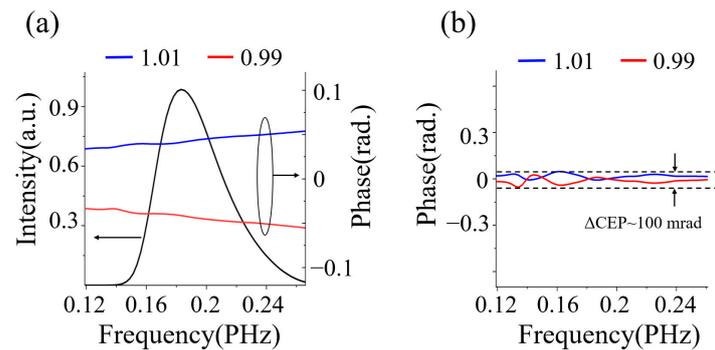
**Figure 3.** The idler spectrum (black solid line) and phase-change curves (a), and CEP-change curves (b) with different input intensity (red line—0.99  $I_a$ , red dash line—0.995  $I_a$ , blue line—1.01  $I_a$ , blue dash line—1.005  $I_a$ , where  $I_a$  is the average intensity).

The SST suppression during nonlinear spectrum broadening by intermediate compression between two spectral broadening stages has already been experimentally demonstrated [25]. Here, we propose to suppress the intensity dependent dispersion caused by SST through the same measure. Figure 4 presents the spectrum and phase-change curves of two-stage scheme with 30 fs input. The thickness of fused silica is 0.3 mm for both stages. After the first stage, the pulse is compressed with GDD of  $-60 \text{ fs}^2$ . Before entering the second stage, the light pulse is attenuated to keep the same peak intensity as the first stage input, which can be realized by magnifying the beam size. The spectrum now is much more symmetric compared to the single stage case (Figure 1) because the SST is much weaker. Meanwhile, the spectral components marked by the light grey rectangles have quasi-linear phase-change curves, which means the intensity dependent dispersion is greatly mitigated.



**Figure 4.** Input (green solid line), broadened (black solid line) spectra, and phase-change curves with different input intensity (red line—0.99  $I_a$ , blue line—1.01  $I_a$ , where  $I_a$  is the average intensity) for two-stage scheme.

The idler spectrum and phase-change curves after DFG are presented in Figure 5a. The spectrum spans from 0.14 PHz to beyond 0.25 PHz. The spectral components are weaker at red side and stronger at blue side compared to the single stage case because of the different broadened spectral shapes. It is obvious that the CEP fluctuation is decreased effectively. It is less than 100 mrad throughout the whole spectral range, which is not only favorable for ultra-stable CEP, but also beneficial for high-precision CEP measurement.



**Figure 5.** The idler spectrum (black solid line) and phase-change curves (a), and CEP-change curves (b) with different input intensity (red line— $0.99 I_a$ , blue line— $1.01 I_a$ , where  $I_a$  is the average intensity).

#### 4. Discussion

CEP stable few-cycle and single-cycle pulses are desired for many laser-matter interaction researches. And for some of them, extreme low CEP noise is needed. For instance, it is showed that stabilizing the beam pointing of the relativistic electrons generated by plasma accelerator driven by near-single-cycle pulse below 1 mrad requires CEP noise less than 50 mrad [26]. To obtain such pulses at various wavelength, IP-DFG of the spectral broadened pulses is the most feasible approach. With this approach, it is crucial to suppress the intensity dependent dispersion during spectral broadening process unless the intensity noise of the laser is negligible, which is invalid for Ti:sapphire lasers. In recent years, the high repetition rate CEP stable light sources driven by ps or 100 s fs pulses of  $1 \mu\text{m}$  have gained more and more attention. Such pulses can be provided with Yb:KGW lasers and Yb:YAG thin disk lasers, which have lower intensity noise compared to Ti:sapphire lasers. However, their pulse duration is much longer than that of Ti:sapphire lasers, thus larger nonlinearity is requested for the spectral broadening process. Therefore, the suppression of intensity dependent dispersion is still compulsory. As aforementioned, the intermediate compression during spectral broadening stages has been employed to suppress the SST. But in this work, it is proposed that the intermediate compression may provide an effective way for suppressing the intensity dependent dispersion. For applications requesting extreme low CEP noise or with very noisy input pulses, multiple compression stages can be used without dramatically increasing the complexity of the system. For instance, the usage of three intermediate compression stages have been reported to achieve single-cycle pulses with input from a Yb:KGW laser [27].

#### 5. Conclusions

In summary, our theoretical study of nonlinear spectral broadening shows that the intensity noise for nonlinear spectral broadening can introduce oscillations of the phase-change curves at the central part of the spectra, while phase-change curves are smoother for the spectral components beyond the outmost peaks. We propose to obtain well-phased spectral components at the wings by intermediate pulse compression, so as to substantially reduce the intensity noise induced dispersion caused by SST. The effectiveness of our proposal has been verified by simulations for input pulse durations of 30 fs. The output pulses are extremely suitable for generation of CEP stable few-cycle sources for laser-matter interaction researches. Although spectral broadening based on multiple thin-plates scheme is considered here, the conclusion is applicable for other spectral broadening techniques.

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