Investigation of the superfluorescence and signal amplification in an ultrabroadband multiterawatt optical parametric chirped pulse amplifier system

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Abstract. We report on theoretical and experimental investigation of the signal and superfluorescence amplification in a 7 TW optical parametric chirped pulse amplifier (OPCPA) system. We demonstrate that the compressed pulse contrast ratio can be improved by using larger seed pulse energies and by careful adjustment of the pump intensity in each amplification stage. Comparison of the measured amplified signal and superfluorescence energies yielded good agreement between calculations and experiments.

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1. Introduction

Since the advent of the optical parametric chirped pulse amplification (OPCPA) many systems based on this technique have been developed [1, 2]. Gain of more than $10^6$ can be achieved over more than 110 THz bandwidth in a few millimetre long amplification region in a single pass [3, 4]. However, this gain is attainable only by using high pump intensities in the amplifier crystal which in turn increases the probability of an efficient parasitic generation and amplification of optical parametric fluorescence (AOPF, or superfluorescence) and deterioration of the pulse contrast ratio.

In a multiterawatt amplifier, the pulse contrast ratio is of major concern because many high-field experiments require clean pulse fronts. The pulse front in an OPCPA is free of pre-pulses as shown in previous measurements [5] but consists of a large pedestal of incompressible superfluorescence which can reach considerable intensities. In optical parametric amplifiers superfluorescence originates from a quantum effect known as optical parametric fluorescence, i.e. spontaneous downconversion of the pump photons into an idler and signal photon pair [6, 7]. As parametric amplification and fluorescence depends on an instantaneous and directional interaction, superfluorescence is produced within the time window defined by the pump pulse. In ultra-broadband OPCPA, there are several ways to suppress superfluorescence: (i) a decrease of parametric gain while sufficient output energy is maintained, (ii) change the temporal overlap between the pump pulse and stretched seed obtaining the best overlap at the pump pulse leading edge, (iii) tight spatial filtering between the amplification stages. In our previous study [5], we showed that shifting the relative delay between the pump and the seed by approx. 30 ps towards the pump leading edge improves the contrast by one order of magnitude. However, detailed study of the parametric gain control in a multi-stage OPCPA system as well as effective suppression of superfluorescence by spatial filtering is still missing.

In this paper, we investigate both theoretically and experimentally the amplification of superfluorescence and the signal in a three-stage OPCPA-system. The main objective of this study is to find a relationship between the pump intensity at each OPCPA stage and the compressed pulse contrast ratio.
Furthermore our study revealed that pump intensity is not the only factor affecting the pulse contrast deterioration. The saturation of an OPCPA stage and angular detuning of the amplifier crystal also contributes to the decreased contrast. In the few-femtosecond range deterioration of the compressed pulse contrast ratio is expected due to sharp features in the spectral edges of the amplified signal [8].

2. Experimental set-up

The optical set-up is shown in figure 1. The OPCPA is seeded by 60% (2.4 nJ) of the output of a Kerr-lens mode-locked Ti:sapphire oscillator. 5.5 fs pulses from the oscillator were stretched to approximately 40 ps using a negative dispersion stretcher [3]. This amount of seed stretching ensures that the intensity of the pump pulse is adequately high over the time window in which all spectral components of the seed pulse are contained. These components are then simultaneously amplified in the OPCPA crystal. Due to losses in the stretcher and low diffraction efficiency (∼3%) of the (AODF, or DAZZLER [9]) the seed energy for the first amplification stage was only 20 pJ. The spectral distribution of the stretched seed pulses is presented in figure 2(a) as the shaded contour. The remaining 40% (1.4 nJ) of the oscillator output was used for all-optical synchronization of the picosecond Nd:YAG pump laser with the Ti:sapphire oscillator. The seed radiation at 1064 nm wavelength was produced in a photonic crystal fiber by soliton-based frequency shifting [10]. Several picojoule of seed energy are obtained and subsequently amplified to 1.4 J in four Nd:YAG amplifiers. After frequency doubling in a Type-II DKDP crystal, the pump laser delivers about 750 mJ at 532 nm pulses with an 8th order supergaussian spatial profile and a nearly Gaussian temporal profile. The pump pulse duration is 80 ps and the pump pulse energy stability at 523 nm is 1.5% rms.

Signal amplification occurs in three Type-I single-pass stages using 5 mm thick ‘AR’-coated BBO-crystals with a noncollinear angle of 2.3°. The OPCPA pump intensities were 18.4, 7.4 and 4.5 GW cm⁻² for the first, second and third amplification stage, respectively. Pump intensity at each amplification stage could be varied independently and the energy was measured using an OPHIR energy meter PE50BB-DIF. We measured the pump pulse energy by averaging over 100 laser shots. Typically the signal pulse is amplified to approx. 90 μJ in the first stage, to approx. 1.8 mJ in the second stage and to 100 mJ in the third stage. The amplified signal energy was measured using OPHIR energy-meter heads PD10, PE10, and PE50BB-DIF depending on expected pulse energy. For measurement of the energy of superfluorescence background at

Figure 1. Optical layout of the three-stage OPCPA; AOPF; acousto-optical dispersive filter (AODF).
each OPCPA stage, the seed was blocked after the pulse stretcher. Measurement of the seed and superfluorescence energy was performed by averaging over 200 laser shots. The measured amplified signal pulse stability was better than 5% rms at the maximum output energy. The spectrum of the amplified pulse is shown in figure 2(curve b). It has a sharp edge near 725 nm because of a steeply rising phase mismatch as can be seen from the calculated parametric gain curve for a 5 mm thick BBO crystal (figure 2(c)). By negative chirping of the signal pulses final compression could be performed as a combined bulk material compressor consisting of 160 mm thick SF57 and 100 mm thick fused silica blocks and a compressor consisting of four positive dispersion chirped mirrors (∼100 fs² bounce⁻¹). The total throughput of the pulse compressor was close to 95%. The residual spectral phase of the compressed pulses was measured directly with a SPIDER interferometer and used as a feedback signal for the DAZZLER for further phase correction. The pulse duration measured with SPIDER was checked using a Grenouille from SwampOptics. Both methods yield approximately the same pulse duration of 14 fs, corresponding to a peak intensity of over 7 TW.

3. Numerical modelling

We have modelled our OPCPA system numerically using a symmetrized Split-Step type method [11, 12]. In this approach, we handle the parametric amplification process in the time domain (nonlinear step). The walk-off effect due to the noncollinearity of the Type-I parametric process is taken into account by shifting the respective field at each loop-step in the direction given by the noncollinear process by adding a first-order term to the spatial phase in equation (4). Higher order dispersion and diffraction terms can also be accommodated by adding the corresponding spectral and spatial phase terms in the Fourier domain (equations (3) and (4)) (linear step). This
becomes necessary if the pulse duration is short (dispersion effects) or in the case of very small beam sizes (diffraction effects).

It is important to note that temporal walk-off due to group-velocity mismatch can usually be neglected in our case because of the long pulses (50 ps) used and the diffraction effects were not important owing to their weak relative impact for unfocused beams. However, for the correct description of superfluorescence spatial effects such as the spatial walk-off for the amplification of fluorescence in three-wave mixing processes are important [13].

The data matrix used in the simulation consists of an array

\[ \text{data}_i = [-R, R] \times [0, T], \]

for each wave (signal, superfluorescence in the signal direction, idler, superfluorescence in the idler direction and pump). The factor R is the radial limit of the beam in the spatial domain which is usually more than three times larger than the FWHM diameter of the pump beam. T is the temporal simulation window which is three times the pump pulse duration at FWHM. In the simulation, we use a matrix with a grid size of 80 \( \times \) 80 and 1000 split-step iteration loops. This choice of parameters represents a compromise between calculation resolution and time required for computation as we used a PC to carry out the simulation.

3.1. Nonlinear step

Since the phase-mismatch (\( \Delta k \)) for the amplified signal and superfluorescence differs, the only way to take into account both amplified field pairs is to simulate the amplification of both fields in interchangeable steps using the same pump intensity. Phase matching effects are included for signal and idler beams by estimating the frequency dependent \( \Delta k \) values in the time domain. The spectral components of the seed in the first stage range from 675 to 1050 nm, which is the acceptance bandwidth of the stretcher (figure 2(a)). In the simulation of the subsequent stages, the spectral span of the signal and the superfluorescence in the signal direction are inherited from the previous stage. The \( \Delta k \) for the superfluorescence is assumed to be zero. This approximation is believed to be consistent since the rate of transition from a pump photon to a signal and an idler photon is higher in the direction of the smallest phase-mismatch and only in that case the OPF can be efficiently amplified from the quantum noise level.

The presence of the OPF is accounted by adding the quantum noise field terms into the coupled wave equations [13, 14]. Therefore, the nonlinear coupled wave equations can be written in the following form:

\[
\frac{\partial A_1}{\partial z} + \alpha_{1m} \frac{\partial A_1}{\partial t} + \cdots + j\beta_{1n} \frac{1}{r} \frac{\partial A_1}{\partial r} \left( r \frac{\partial A_1}{\partial r} \right) + \cdots + \gamma_1 \frac{\partial A_1}{\partial r} = j \kappa_1 A_2^* A_1 e^{-\Delta k_1 z} + \sqrt{\epsilon_1} \xi_1(z, t),
\]

\[
\frac{\partial A_2}{\partial z} + \alpha_{2m} \frac{\partial A_2}{\partial t} + \cdots + j\beta_{2n} \frac{1}{r} \frac{\partial A_2}{\partial r} \left( r \frac{\partial A_2}{\partial r} \right) + \cdots + \gamma_2 \frac{\partial A_2}{\partial r} = j \kappa_2 A_1^* A_2 e^{-\Delta k_2 z} + \sqrt{\epsilon_2} \xi_2(z, t),
\]

\[
\frac{\partial A_3}{\partial z} + \alpha_{3m} \frac{\partial A_3}{\partial t} + \cdots + j\beta_{3n} \frac{1}{r} \frac{\partial A_3}{\partial r} \left( r \frac{\partial A_3}{\partial r} \right) + \cdots + \gamma_3 \frac{\partial A_3}{\partial r} = j \kappa_3 A_1 A_2 e^{\Delta k_3 z} + \sqrt{\epsilon_3} \xi_3(z, t),
\]

where \( \kappa_l \) are the nonlinear coupling coefficients [15] calculated for the central frequencies, \( A_l \), \( l = 1 \ldots 3 \) represent the normalized complex field amplitudes for the signal (or the...
superfluorescence field in the signal direction), idler (or the superfluorescence field in the idler direction) and pump fields, respectively. For the description of the noise fields, we will use the approach first introduced by Gatti et al [14]. The complex stochastic variables $\xi_l(z, t)$ have a Gaussian distribution with a zero mean value $\langle \xi_l(z, t) \rangle = 0$ and the correlation $\langle \xi_l(z, t) \xi_j^*(z', t') \rangle = \sigma_l \sigma(t - t') \sigma(z - z')$. Here $\epsilon_i$ are the noise intensities of the respective fields. A similar approach was used to describe the influence of temporal and spatial walk-off during the parametric amplification of stochastic fields [13]. The intensity $\epsilon_3$ is set to zero as the pump field is already initialized with the complex amplitude $A_3$. $\alpha_{im}$ are the dispersion coefficients with order $m$ whereas $\beta_{in}$ are the diffraction coefficients with order $n$. $\gamma_i$ is the walk-off coefficient due to the noncollinear geometry of interaction. This coefficient for the pump ($\gamma_3$) is in our case zero as the pump propagates normal to the crystal plane.

3.2. Linear step

Dispersion effects are taken into account in the frequency domain [16]. $\Delta \varphi_r(\omega)$ is the spectral phase expanded in a Taylor series.

$$A_l(r, z + \Delta z, t) = \mathcal{F}^{-1}\{\mathcal{F}\{A_l(t, z)\} e^{i\Delta \varphi_r(\omega)}\}. \quad (3)$$

$\mathcal{F}$ is the Fourier transformation and $\mathcal{F}^{-1}$ is the inverse Fourier transformation.

The effects of diffraction and the walk-off due to the noncollinearity of the type I parametric process is taken into account by adding a spatial phase term $\Delta \varphi_r(\omega) = \Delta \varphi_{nc}(\omega) + \Delta \varphi_{diff}(\omega)$ (the subscripts nc and diff correspond to the noncollinear and diffraction terms respectively) in equation (4) by applying the same Fourier (linear) step to the transposed $(-R, R) \times (0, T)$ data matrices $A_l^\dagger(r, z + \Delta z, t)$.

$$A_l^\dagger(r, z + \Delta z, t) = \mathcal{F}^{-1}\{\mathcal{F}\{A_l^\dagger(t, z)\} e^{i\Delta \varphi_r(\omega)}\}. \quad (4)$$

To improve agreement between simulation results and experiments, the noise intensities $\epsilon_i$ have to be found empirically for the first simulation run. One possibility to estimate the noise intensities is to measure the superfluorescence in absence of the seed, simulate the system with the same pump intensity and tune the input noise intensities such as to obtain the same superfluorescence output as in the experiment. This method is reliable if the ratio between amplified signal and superfluorescence is large and for the amplifier stage far from saturation.

4. Simulations versus experimental results

This code was used to calculate amplification of the signal pulse and the AOPF in the three-stage OPCPA system described in section 2. It is important to mention here that after the pulse compressor the contrast ratio between the peak intensity of amplified pulse and instantaneous intensity of superfluorescence for our three-stage OPCPA is estimated to improve by a factor approximately equal to the compression ratio of approx. $2 \times 10^3$. Thus superfluorescence level of 100% from the amplified signal energy after the compression causes pulse contrast ratio of approximately four orders of magnitude. This result is consistent with our previous measurement results using a high-dynamic range third order correlator [5].
4.1. Deterioration of the contrast in the first amplifier stage

The energy evolution in the first amplification stage of the amplified signal and superfluorescence versus the pump intensity is depicted in figure 3(a). It is evident that the experimental points show agreement with the calculations. The superfluorescence in the first stage has much more favourable phase matching conditions than the seed due to the angular dispersion of its frequency components and due to the fact that it has the same amount of downconverted signal and idler photons from the moment of generation (after propagation through the critical length for parametric generation in the crystal [17]). In contrast, the signal needs a longer propagation distance to build-up a comparable amount of idler photons for the parametric amplification. In our case the signal after the propagation through this critical length is approximately $10^3$ times stronger than the superfluorescence. The best pulse contrast ratio that can be obtained from this OPCPA stage is $4 \times 10^{-6}$ if the signal is compressed to the TL as shown from simulation results (figure 3(b)). To explore the connection between seed energy and temporal pulse contrast,
The simulation of the first stage is carried out by varying seed energy. This plot is not valid in the case of the saturated amplification. The pulse contrast ratio is simulated numerically for an input seed energy varying from 0.5 to 50 times the current seed energy \( E_{10} \) of 20 pJ. We conclude that superfluorescence deteriorates the pulse temporal contrast ratio in the first stage as a consequence of the low seed energy in combination with the high pump intensities used in this stage to obtain high gain of the order of \( 10^5 \). Therefore, a straightforward way to increase the pulse contrast ratio in an OPCPA would be to develop a stronger seed source. However, this cannot lead to arbitrary improvement of pulse contrast because, as we can deduce from the results presented in figure 3(b), it does not make much sense to use seed energies bigger than 10 nJ because the final limitation for such seeding is the contrast of the ultrabroadband oscillators (in the \( \pm 5 \) ps temporal window) [18, 19] marked as a shadowed area in figure 3(b). To improve pulse contrast beyond oscillator contrast the nonlinear pulse cleaning methods such as the cross-polarized wave generation (XPW) [20] or the plasma mirror [21] which were developed for use with convention CPA systems could be used. Unfortunately, both techniques are difficult to implement and result is reduced output energy with a tremendous increase of the set-up complexity.

4.2. Influence of the angular detuning

The energy evolution in the second amplification stage of the amplified signal and superfluorescence versus the pump intensity is depicted in figure 4(a). For the numerical code, we used 90 \( \mu \)J and 850 nJ input energies for the signal and superfluorescence, respectively. These energies are obtained using approx. 18 GW cm\(^{-2}\) pump intensity in the first stage (figure 3(a)). From this data, we can conclude that there are noticeable discrepancies between experimentally measured and calculated signal energies. This could be explained if we remember that the second stage in our set-up is not only used to reach the millijoule-level but also to favour the amplification of the spectrum edges to compensate for the smaller amplification in this region. Slight angular detuning in the second OPCPA stage allows us to reshape the gain bandwidth [22] and reduce the amplification bandwidth narrowing throughout the amplifier. This causes lower gain for the signal due to increased phase mismatch. Illustration of gain reshaping due to angular detuning is presented in figure 4(b) where experimentally measured superfluorescence spectra for the exact phase-matching (shaded area) and for \( \Delta k \neq 0 \) (solid line) are plotted. Therefore, we attribute this discrepancy between experimental and calculated results to the difference between the \( \Delta k \) values used in the experiment and in the numerical code. For the calculation of the superfluorescence amplification, we again assumed perfect phase matching (\( \Delta k = 0 \)).

Another important experimental observation is that the pulse contrast does not deteriorate much in the 2nd stage. This result can be attributed to the common action of the low gain (~20) and angular detuning. The observed energy difference and the known compression ratio allows us to estimate the pulse contrast in the current system to be approx. \( 2 \times 10^{-5} \) for the 2.3 mJ pulses. Previously, the pulse contrast ratio for similar pulse energies in a two stage OPCPA was measured after compression to approx. 10 fs pulse duration [5]. The best result obtained was approx. \( 5 \times 10^{-5} \).

4.3. Influence of the gain saturation

In the last amplifier stage of our system saturation is reached for the signal (figure 5(a)). In the simulation, we used 1.8 mJ and 100 \( \mu \)J input energies for the signal and superfluorescence,
Figure 4. (a) Dependence of the amplified signal and superfluorescence energies on the pump intensities in the second stage; (b) measured superfluorescence spectra in the second stage for two different phase matching conditions.

respectively. These energies are obtained using approx. 7 GW cm\(^{-2}\) pump intensity in the second stage (figure 4(a)). Gain saturation improves the amplified signal energy stability which was 5% rms obtained with a pump energy stability of less than 2%. As already mentioned in the experimental set-up description the usual procedure to measure the superfluorescence in an OPCPA stage is to block the seed in front of the first stage. However, in the absence of the seed, the pump energy is used only for the superfluorescence amplification and as a consequence the superfluorescence will be amplified to higher energy than in the presence of the signal. In the first two stages, this effect is not so pronounced as the difference between signal and superfluorescence energies is substantial but in the last amplifier stage the pump and amplified signal energies become comparable. For that reason in the third stage, we performed calculations of the superfluorescence amplification with and without presence of the seed (figure 5(a) superscripts \(^\ast_1\) and \(^\ast_2\), respectively). It is evident that the presence of the seed causes parametric gain quenching [23]. For a gain of 50, the ratio between the signal and superfluorescence energies is about 1 : 7 instead of 1 : 2 as shown in figure 5(a)). Assumption of the presence of the gain quenching in the saturated OPCPA stage allowed us to calculate the temporal contrast ratio of the 7 TW OPCPA
5. Conclusions

We explored the signal and superfluorescence amplification in a multistage few-cycle OPCPA system by comparing experimental data and those generated by our numerical model. This provided insight into various problems related to pulse contrast which is a crucial issue in high-field physics experiments. From our study, we conclude that the pulse energy from a Kerr-lens modelocked Ti: sapphire oscillator is too low to seed the multiterawatt OPCPA system presented here. Another more encouraging result is that gain saturation does not substantially deteriorate the temporal pulse contrast ratio due to the parametric gain quenching-effect in the presence of the signal wave. There are several possibilities for a contrast improvement. Two of them have been discussed in the previous section as the XPW and the plasma mirror. From the data obtained in the second and the third OPCPA stage, it is evident that to avoid superfluorescence generation...
the parametric gain should be kept at about $10^2$–$10^3$. The pulse contrast measurements and further contrast enhancement using tight spatial filtering of the three-stage OPCPA system are under way.

References

[1] Dubietis A, Jonusauskas G and Piskarskas A 1992 Opt. Commun. 88 437
[2] Dubietis A, Butkus R and Piskarskas A 2006 IEEE J. Sel. Top. Quantum Electron. 12 163
[3] Ishii N et al 2005 Opt. Lett. 30 567
[4] Witte S, Zinkstok R Th, Hogervorst W and Eikema K S E 2005 Opt. Express 13 4903
[5] Tavella F, Schmid K, Ishii N, Marcinkevicius A, Veisz L and Krausz F 2005 Appl. Phys. B 81 753
[6] Kleinman D A 1968 Phys. Rev. 174 1027
[7] Danielius R, Piskarskas A, Stabinis A, Banfi G P, Di Trapani P and Righini R 1993 J. Opt. Soc. Am. B 10 2222
[8] Osvalt K, Csatari M, Ross I N, Persson A and Wahlström C G 1993 Laser Part. Beams 23 327
[9] Verluise F, Laude V, Cheng Z, Spielmann Ch and Tournois P 2000 Opt. Lett. 25 575
[10] Teisset C Y, Ishii N, Fuji T, Metzger T, Köhler S, Holzwarth R, Baltuska A, Zheltikov A M and Krausz F 2005 Opt. Express 13 6550
[11] Kurtanaitis A, Dementjev A and Ivanauskas F 2001 Nonlinear Anal.: Model. Control 6 51
[12] Arisholm G 1997 J. Opt. Soc. Am. B 14 2543
[13] Picozzi A and Haelterman M 2001 Phys. Rev. E 63 0566111
[14] Gatti A, Wiedemann H, Lugiato L A, Marzoli I, Oppo G and Barnett S M 1997 Phys. Rev. A 56 877
[15] Reider G 1997 Photonik (New York: Springer)
[16] Ross I N, Matousek P, New G H C and Osvalt K 2002 J. Opt. Soc. Am. B 19 2945
[17] Carrion L and Girardeau-Montaut J 2000 J. Opt. Soc. Am. B 17 78
[18] Kasper A and Witte K J 1998 J. Opt. Soc. Am. B 9 327
[19] Braun A, Rudd J V, Cheng H, Mourou G, Kopf D, Jung I D, Weingarten K J and Keller U 1995 Opt. Lett. 18 2490
[20] Cotet A, Jullien A, Forget N, Albert O, Cheriaux G and Le Blanc C 2006 Appl. Phys. B 83 7
[21] Doumy G, Quere F, Gobert O, Perdrix M, Martin Ph, Audebert P, Gauthier J C, Geindre J-P and Wittmann T 2004 Phys. Rev. E 69 026402
[22] Sosnowski T S, Stephens P B and Norris T B 1996 Opt. Lett. 21 140
[23] Kondo K, Maeda H, Hama Y, Morita S, Zoubir A, Kodama R, Tanaka K A, Kitagawa Y and Izawa Y 2005 J. Opt. Soc. Am. B 2 231