Design, Optimization, and Reverse Engineering of High-Power Laser Systems

Jack Hirschman$^{1,2,*}$, Randy Lemons$^{2,3}$, Minyang Wang$^4$, Peter Kroetz$^5$ and Sergio Carbajo$^{2,6,7}$

$^1$Department of Applied Physics, Stanford, 348 Via Pueblo, Stanford, CA 94305, USA.
$^2$SLAC National Accelerator Laboratory, 2575 Sand Hill Rd, Menlo Park, CA 94025, USA.
$^3$Colorado School of Mines, 1500 Illinois St, Golden, CO 80401, USA.
$^4$Statistics Department, UCLA, 520 Portola Plaza, Los Angeles, CA 90095, USA.
$^5$Atomically Resolved Dynamics Division, Max Planck Research Department for Structural Dynamics, University of Hamburg, 20146 Hamburg, Germany.
$^6$Electrical and Computer Engineering Department, UCLA, 420 Westwood, Los Angeles, CA 90095, USA.
$^7$Physics and Astronomy Department, UCLA, 475 Portola Plaza, Los Angeles, CA 90095, USA.

*Corresponding author(s). E-mail(s): jhirschm@stanford.edu;

Abstract

Chirped pulse amplification (CPA) followed by nonlinear optical (NLO) systems constitute the backbone of myriad advancements in semiconductor and additive manufacturing, communication networks, biology and medicine, defense and national security, and a host of other sectors over the past decades. However, accurately and efficiently modeling CPA- and NLO-based laser systems is challenging because of the multitude of coupled linear and nonlinear processes and high variability in modeling frameworks. The lack of fully-integrated software CPA+NLO modeling severely hampers further advances to tailor existing or materialize new CPA+NLO systems and curtails their full potential in emerging inverse...
design approaches through data-driven machine learning methods. Here, we present a modular start-to-end software model, allowing for the inclusion of an array of amplifier designs and nonlinear optics techniques, which renders time- and frequency-resolved electromagnetic fields alongside essential physical characteristics on energy, fluence, and spectral distribution. To demonstrate its robustness and real-world applicability—specifically, reverse engineering, system optimization, and inverse design—we present a case study on the LCLS-II photo-injector laser, representative of a high-power and spectro-temporally complex CPA+NLO system.

**Keywords:** CPA, software model, NLO, reverse engineering

## 1 Introduction

The 2018 Nobel Prize in Physics was partially awarded to Donna Strickland and Gerard Mourou for their monumental work on chirped pulse amplification (CPA) of optical pulses [1, 2], which they first published on in their seminal piece in 1985, _Compression of Amplified Chirped Optical Pulses_ [3]. Since then, CPAs have driven and rapidly expanded the fields of high-power lasers, ultrafast lasers, and nonlinear optics (NLO) and now touch every realm of our lives with applications in cancer treatment, precision machining in semiconductor and electronics manufacturing, and surgery among the countless other applications. Nevertheless, ground-up design of high power CPA systems continues to be non-trivial, especially for those setups involving cascaded nonlinear subsystems. Choosing the correct hardware and determining limitations for a particular laser system—such as damage thresholds for optics and amplifiers as well as bandwidth limitations for shaping—can be a challenging task, especially for systems involving custom design amplifiers, pre-CPA programmable pulse shapers, or a series of up-/down-conversion stages. Start-to-end (S2E) software models can greatly inform laser system design decisions and aid in understanding limitations or trade-offs in performance parameters. To an even greater extent, as we enter new stages of machine learning (ML)-driven photonics and optical system design [4], the need for S2E-based data generation to inform these ML models and reduce parameter searches in the lab will become increasingly desired. Accurate, robust, and comprehensive software models can not only catalyze the ground-up design of CPA systems and cascaded nonlinear conversion but can also help find new paradigms in the science, engineering, and applications of ultrafast laser pulses.

Of course theory and models for certain subsets of CPA systems have existed for a some time. Frantz and Nodvik with their 1963 work on pulse propagation in a laser amplifier via the photon transport equations [5] laid the groundwork for much of the modern theory, simulation, and texts regarding pulse amplification [6, 7]. For nonlinear optical systems and pulse propagation through fibers and waveguides or free-space, the simulation largely
follows along either the unidirectional pulse propagation equation (UPPE) developed by Kolesik, Moloney, and Mlejnek [8, 9] or the generalized nonlinear Schrödinger equation (GNLSE). The GNLSE was developed in stages where first a method based on the nonlinear Schrödinger equation was used to describe pulse propagation in optical fiber [10] and derived via the slowly varying envelope approximation (SVEA). Brabec and Krausz used the slowly evolving wave approximation (SEWA) to then build what has become known as the GNLSE [11], which of course contains the mixed derivatives of the time- and spatial-coordinates as well as the second-order time derivative [12]. Modeling these equations has required development in accompanying numerical solvers. In general, the carrier resolved propagation equations like UPPE, forward Maxwell equation, and others can be solved by spectral methods while nonlinear envelope propagation equations such as the GNLSE require a combination of finite-difference and spectral methods [13]. The numerical backbones like finite-difference time domain [14], split-step Fourier method, Runge-Kutta, and many others have been heavily used and improved on for simulation in nonlinear optics and pulse propagation. Furthermore, there are sophisticated software packages available for modeling particular phenomena in fiber and free space including those by the Agrawal group [15], the Travers group [16], and others. However, a fast start-to-end software model for ground-up design of CPA systems involving cascaded nonlinear sections still does not exist.

Such a fully encompassing model built up from first principles is difficult for several reasons. First, such a system includes many cascaded nonlinear processes, including both literal nonlinear optical processes and also nonlinear equations governing standard processes. Next, while models for various subsystems have existed in isolation, interfacing these in a way that is computationally efficient and physically correct requires care. Finally, such a model must be robust enough to be able to incorporate new modules and be able to interface with other simulators in a way to further increase its applicability, such as feeding into a particle acceleration simulator. While such a S2E model would be complex, the application space would be immense with sectors ranging from defense and national security to biomedicine all benefiting from ground-up design of CPA and NLO systems, generating data for ML for these systems, or even reverse engineering such systems.

Here, we present this S2E model that addresses the need for modeling integrated systems that involve shaping, amplification, and up-/down-conversion stages. Furthermore, our design is both modular and customizable such that one can tailor it to specific applications. In this paper, we will first provide a more detailed motivation for developing the model, showing examples from many sectors that would require CPAs and that would benefit from such a S2E model. Then we describe the physical and mathematical foundations of key elements in the S2E model. Finally, to demonstrate the model’s robustness, we take the photo-injector laser at SLAC National Laboratory’s LCLS-II, the world’s most powerful XFEL, as an example use case. The photo-injector laser produces the ultraviolet (UV) pulses used for generating the electrons, which
are accelerated, shaped, and ultimately undulated to produce femtosecond and attosecond coherent X-ray pulses.

Figure 1 shows the main building blocks for this photo-injector laser, which starts with a mode-locked infrared (IR) oscillator which undergoes spectral phase and amplitude shaping, followed by amplification via a Yb:KGW regenerative amplifier (RA), and finally upconversion to UV. In our case study, we use noncolinear sum-frequency generation (SFG) followed by second harmonic generation (SHG) [17]. We use our adaptable software model to match the model of the amplifier to the physical system. This example thus allows us to show how our S2E model can be used for tuning and matching the model to a physical amplifier via reverse engineering, how the model can be used for optimizing amplifier parameters for ground-up design or adjustments, and how the model can be used for inverse design of materials. Finally, this example also will be used to demonstrate the modeling of cascaded processes by inspecting combinations of pulse shaping, CPA, and upconversion.

2 Motivation

CPA and NLO systems have an extremely far-reaching application space, and the multitude and diversity of stakeholders in such a S2E software to model these systems are what motivate this development. These stakeholders span many sectors, and it is not practical to list each application specifically, but it is important to give an idea of whom such a S2E model could assist in designing systems from the ground-up, reverse engineering current systems to enable better software-hardware harmony, or enabling ML-based inverse design.
In the defense and national security sphere, applications involve atmospheric feedback and shaping, spectroscopy for materials identification, remote sensing, target tracking, and direct energy weapons systems. For example with remote sensing and LIDAR, CPA was proposed as a solution to achieve the pulse durations and intensities [18], and now more recently optical parametric CPA (OPCPA) is often used to generate the required high power laser pulses [19]. Building on these techniques, LIDAR has been adapted to remote sensing of trace gases in the atmosphere, such as by the use of optical correlation spectroscopy LIDAR (OCS-LIDAR), which uses an acoustic optic programmable dispersive filter (AOPDF) to perform spectral shaping of the laser pulse to tune it to the gas of interest [20]. Similarly CPAs are required in studies of filamentation and propagation of high-intensity, ultra-short pulses in air. For instance, the air will experience breakdown at high enough intensities, leading to the creation of a plasma. The effects of the plasma on the index of refraction can actually balance each other and lead to noncollapsing filaments, which can undergo extremely long propagation. At intensities on the order of $10^{13}$ W/cm$^3$ or higher, secondary radiation can be generated which disrupts the operation of electronics and thus shows to be a possible application for sensor damage and electronic countermeasures [21]. When partnered with work showing that filamentation can extend kilometers in distance [22, 23] and transmit through clouds [24] and fog [25], the potential uses for these systems as successful directed-energy instruments really shines [26]. Building on these advancements, opportunities for using femtosecond pulses and induced filamentation for secure free-space communication networks opened up. Furthermore, pulse shaping was introduced pre-amplification in order to compensate for changing atmospheric conditions by using a genetic algorithm to feed updated parameters to an AOPDF to perform adaptive corrections [27].

In basic research sector, the applications are extremely expansive covering many areas in high energy physics including initial particle generation for accelerators and particle acceleration. For instance, laser wakefield acceleration heavily relies on CPA-based systems to achieve a sufficiently high laser intensity to expel electrons from the blow-out region where the electron beam can propagate and be accelerated. Now that the field is maturing, pulse shaping devices are being incorporated into feedback loops for corrections on these lasers to further improve the electron beam quality [28]. For free-electron lasers (FELs), the temporal shape of the photo-injector laser affects the emittance and other characteristics of the electron distribution at the interaction point with the photocathode. Recent studies are exploring how to alter these temporal shapes of the photo-injector laser in order to reduce initial electron emittance and even drive microbunching in the electron bunch in order to stimulate attosecond X-ray pulses in X-ray free-electron lasers (XFELs), for instance [17, 29, 30]. Then for Compton scattering, for example, recent efforts at improving photon density for Compton scattering by modulating the incident laser have proposed using flat-top profiles [31] and linear chirp to reduce the ponderomotive broadening. Recently, using catastrophe theory and
numerical simulations, it was shown that using linear chirped lasers increases photon yield [32]. For high harmonic generation, the effects of pulse shaping on the upconverted harmonic spectrum have been studied for some time. For example, Chang, et al. introduced spectral chirp to investigate temporal phase control of soft X-ray emission [33], and many others have followed on with more advanced shaping techniques since then [34, 35]. For inertial confinement fusion (ICF), the advent of CPA ushered in new areas of research in ICF by allowing the use of an external ignitor beam for fast ignition [36] and later opened the door to schemes incorporating pulse shaping as well [37].

In the medical and biomedical fields CPA and NLO systems are required in proton acceleration for proton therapies in cancer treatment, dynamical imaging, static imaging, and even fluorescent labeling. For example, the field of 2D IR spectroscopy, researchers have introduced the use of pulse shapers and diffractive optics to ensure precise-time delays while also keeping the measurement more robust [38], and other groups have begun using pulse-shaping pre-IR upconversion using four-wave mixing such that over 100 spectra could be collected in a 1 hr time frame as opposed to just one spectrum in an hour before the introduction of the pulse shaper [39]. And similarly the importance of pulse shaping in nuclear magnetic resonance has been shown for some time ever since the field began transitioning from continuous to pulsed irradiation and subsequently began exploring shapes beyond flat-top rectangular pulses [40]. Ultrafast laser pulses have also had a significant effect on surgery such as in femtosecond-LASIK, dental operations with treatment of carious tissue, ear surgery including reconstruction of defect auditory ossicles, and treating cardiovascular disease such as with extraluminal laser angioplasty among other applications [41].

Finally in industry, applications are extremely far-ranging. For instance, CPAs enabled the field of femtosecond laser machining which can create clean ablation craters with the use of sufficiently intense and short pulses [42]. Similarly, these lasers can create microstructures on solid surfaces in a way that avoids chemical etching [26, 43]. CPA and NLO obviously play a key role in fiber optic communications, and many applications have developed as a result of the fiber optic network lines. For example, fiber-optic distributed sensing (DAS) and chirped-pulse DAS (CP-DAS) have enabled these networks to be used for monitoring oil and gas boreholes, cable integrity, seismic activity, and pipelines by sending short pulses through the fibers and looking at the changes in the internal interference based on the applied strain to the fibers [44, 45]. High intensity, short pulse lasers are also used in advanced materials design. For instance, femtosecond pulses can drive shock compression in graphite to achieve the hexagonal diamond transformation [46].

The application space for CPA and NLO systems continues to expand, and we can see how these systems underpin nearly every area of modern life. Furthermore, we have seen that while there are many sophisticated methods for modeling components of these systems, a fully integrated start-to-end software model incorporating CPA, NLO, pulse shaping, and other functionalities does
not yet exist. Our work presents this missing simulation model and shows results for a model configuration based on the components in LCLS-II’s photo-injector laser.

3 Methods

Our S2E model uses a modular design for flexibility in simulation. Each module takes an input electric field or set of electric fields, input parameters that define how that block operates, an output or set of output electric fields, and a set of peripheral outputs containing additional simulation information. An block diagram of the modules incorporated into the simulator are shown in figure 1 and include a mode-locked oscillator, a pulse shaper, regenerative amplifier, and upconversion techniques including SFG and SHG.

The pulse shaper is shown in figure 1 (b) and is modeled as a transfer function applied to the zero-centered frequency-domain input electric field,

\[ \tilde{E}_{\text{out}}(\omega) = \tilde{E}_{\text{in}}(\omega) \cdot F(\omega) \]  

(1)

where \( F(\omega) \) is the transfer function. The output in time is then the inverse Fourier transform

\[ E_{\text{out}}(t) = F^{-1}\tilde{E}_{\text{out}}(\omega). \]  

(2)

The transfer function consists of an amplitude shaping portion \( A(\omega) \) and a phase shaping portion \( \phi(\omega) \),

\[ F(\omega) = A(\omega) \cdot e^{i\phi(\omega)}. \]  

(3)

The amplitude shaping is a super-Gaussian pulse extending beyond the bandwidth of the input pulse modulated by an inverted Gaussian pulse with tunable depth, width, and peak position.

\[ F(\omega) = e^{-(\omega/\delta\omega_0)^6} \cdot \left(1 - k \cdot e^{-((\omega+\omega_0-\omega_1)/\delta\omega_1)^2}\right) \cdot e^{i\phi(\omega)} \]  

(4)

where \( \omega \) is the angular frequency domain, \( \omega_0 \) is the central frequency of the input electric field, \( \delta\omega_0 \) is the width parameter of input electric field, \( k \) determines the depth of the notch filter—between 0 and 1—, \( \omega_1 \) is the position in frequency of the notch, \( \delta\omega_1 \) is the width parameter for the notch, and \( \phi(\omega) \) is the phase function.

Figure 2 shows this transfer function waveform shape as a black, dashed line. It forms a super-Gaussian with a notch that spans the entirety of the input pulse spectral amplitude (shown in red). Here the notch is centered at 1030 nm with a width of about 10 nm. The input pulse is centered at 1035.5 nm.

The phase transfer function can be an arbitrary function; however, we parameterize it using a Taylor expansion up to fourth-order. This method naturally allows for incorporating first-order dispersion (delay), second-order
dispersion (SOD), third-order dispersion (TOD), and fourth-order dispersion (4OD). Thus, the function can be represented as

$$\phi(\omega) = - \left( a_1 \cdot (\omega) + \frac{a_2}{2} \cdot (\omega)^2 + \frac{a_3}{6} \cdot (\omega)^3 + \frac{a_2}{24} \cdot (\omega)^4 \right)$$  \hspace{1cm} (5)$$

where the terms $a_1$ to $a_4$ correspond to the dispersion orders and the field is assumed to be centered at zero.

Figure 2 also shows the phase portion of the transfer function a black, dashed line. The input field has flat phase, and the transfer function is a mix of SOD and TOD which amounts to quadratic and cubic contributions, respectively.

The final equation, thus, takes the form

$$F(\omega) = e^{-\left(\frac{\omega}{\delta \omega_0}\right)^6} \cdot \left(1 - k \cdot e^{-\left(\frac{\omega + \omega_0 - \omega_1}{\delta \omega_1}\right)^2} \right) \cdot e^{-i \cdot \left( a_1 \cdot (\omega) + \frac{a_2}{2} \cdot (\omega)^2 + \frac{a_3}{6} \cdot (\omega)^3 + \frac{a_2}{24} \cdot (\omega)^4 \right)}$$  \hspace{1cm} (6)$$

where $\omega_0$ only shows in the portion forming the notch so that the notch is placed properly with respect to the recentered carrier frequency.

In figure 1 the CPA follows the pulse shaper. Here we model a regenerative amplifier (RA), specifically the spectral evolution of the input pulse through the amplifier, based on the work of Kroetz, et al [7]. The model generalizes the Frantz-Nodvik [5] equations to work across a spectral bandwidth, which is necessary to inspect chromatic effects in the amplification process as the original equations only handled monochromatic light.
This theory begins with the nonlinear, time-dependent photon transport equation \[5, 6\]
\[
\frac{\partial \phi}{\partial t} + c \left( \frac{\partial \phi}{\partial z} \right) = \sigma c n (N_2 - N_1) \tag{7}
\]
where $\phi(z,t)$ is the photon density, $N_1(z,t)$ is the number density of electrons in the ground state, $N_2(z,t)$ is the number density of electrons on the excited state, $\sigma$ is resonance absorption cross section, $c$ is the speed of light in the material, and $z$ is the direction of propagation through the crystal. The equation shows how the rate of increase of the photon density is related to the flow of flux out of the volume of material and the increase in photons due to stimulated emission.

In a lossless system, the rate of decrease of excited state electrons must balance the rate of increase of ground state electrons such that

\[
\frac{\partial N_1}{\partial t} = \sigma c \phi (N_2 - N_1) \tag{8}
\]
\[
\frac{\partial N_2}{\partial t} = -\sigma c \phi (N_2 - N_1) \tag{9}
\]
where $N_1 + N_2 = N_{Total}$ is a constant.

We can then introduce the population difference $n$ as $n = N_2 - N_1$ yielding

\[
\frac{\partial n}{\partial t} = -\gamma n c \sigma \phi \tag{10}
\]
where $\gamma$ relates to the total reduction in the inverted population after single photon emission. Here, $\gamma = 2$ under the assumption of no degeneracy in the three-level system, but, more generally, can be defined as

\[
\gamma = 1 + \frac{g_2}{g_1} \tag{11}
\]
where $g_1$ and $g_2$ are the degeneracies for the ground and excited states, respectively, and relate to the probability for emission and absorption.

Frantz and Nodvik solved equations 7 and 10 \[5\] for several input pulse cases to the amplifier including a square pulse with duration $t_p$ and initial photon density $\phi_0$. The photon density is given by \[6\]
\[
\frac{\phi(z,t)}{\phi_0} = \left\{ 1 - \left[ 1 - e^{-\sigma n z} \right] e^{-\gamma \sigma \phi_0 c (t - \frac{z}{c})} \right\}^{-1} \tag{12}
\]
and the energy gain for the light passing through the amplifier is

\[
G = \frac{1}{\phi_0 t_p} \int_{-\infty}^{+\infty} \phi(l,t) dt
\]
\[
= \frac{1}{c \gamma \sigma \phi_0 t_p} \ln \left\{ 1 + \left[ e^{\gamma \sigma \phi_0 c t} - 1 \right] e^{n \sigma l} \right\} \tag{13}
\]
where $l$ is the length of the amplifying medium.

Now can formulate this equation in terms of measurable laser parameters. Will start by defining the input fluence as the input energy per unit area which is

$$J_{in} = c \phi_0 t_p h \nu$$

(14)

where $h$ is Planck’s constant and $\nu$ is the central frequency of the input beam. Then can define a saturation fluence as

$$J_{sat} = \frac{h \nu}{\gamma \sigma} = \frac{J_s}{\gamma g_0}$$

(15)

where $J_s = h \nu n$ represents the amount of stored energy in the medium and the small signal gain is given by $g_0 = n \sigma$.

With these expressions, can get an expression for the gain $G$

$$G = \frac{J_{sat}}{J_{in}} \ln \left\{ 1 + \left[ e^{\frac{J_{in}}{J_s}} - 1 \right] G_0 \right\}$$

(16)

where

$$G_0 = e^{g_0 l}$$

(17)

is the small signal, single-pass gain.

Finally we can put this in terms of input and output fluence to the system where

$$J_{out} = J_{sat} \ln \left\{ 1 + \left[ e^{\frac{J_{in}}{J_{sat}}} - 1 \right] G_0 \right\}.$$ 

(18)

For regenerative amplifiers or systems involving multiple passes, the output fluence can be fed back in to the input to cascade the steps.

With these fundamentals established, can begin modifying these monochromatic functions to incorporate spectral information [7]

$$J_{out}(\lambda) = J_{sat}(\lambda) T(\lambda) \ln \left\{ 1 + \left[ e^{\frac{J_{in}(\lambda)}{J_s(\lambda)}} - 1 \right] G_0(\lambda) \right\}. $$

(19)

where we have included $T(\lambda)$ to account for losses in the system. The saturation fluence can be redefined to account for the spectral dependence of the emission and absorption cross sections by using

$$J_{sat}(\lambda) = \frac{hc}{\lambda (\sigma_{abs}(\lambda) + \sigma_{em}(\lambda))}.$$ 

(20)

Now when considering a pulse train and an amplifier with multiple passes of the input laser beam, the energy extracted from the gain medium must be treated carefully. The gain must be updated with each pass of the input pulse and with each pumping period. However, as the work by Ref [7] points out, the gain has spectral dependencies. Instead, Kroetz, et al. suggest updating an inversion fraction, $\beta$, which under the assumption of homogeneous broadening
is wavelength independent. The inversion fraction represents the proportion of excited state ions compared to the total number of ions and is defined as

$$\beta = \frac{N_2}{N_{Total}}.$$  \hfill (21)

Now to account for the multipass nature, the input/output designations can be replaced by subscripts i/i-1, respectively. So the single pass gain from equation 17 can be re-written as

$$G_{i-1}(\lambda) = e^{\sigma_{g,i-1}(\lambda)Nl}$$  \hfill (22)

where the spectral gain cross section is

$$\sigma_{g,i-1}(\lambda) = \beta_{i-1}(\sigma_{em}(\lambda) + \sigma_{abs}(\lambda)) - \sigma_{abs}(\lambda)$$  \hfill (23)

and where $\beta_{i-1}$ is this averaged global inverted fraction and $N$ is the dopant ion density in the material. This inverted fraction at a given step $i$ can be arrived at by considering the update to the gain at a step $i$. In the monochromatic case, we can say that the next value for the small signal gain will be

$$g_i = g_{i-1} + \Delta g_i = g_{i-1} + \frac{\Delta J_{stor,i}}{J_{sat}}$$  \hfill (24)

which first treats the update to the gain based on the change in the stored fluence in the gain medium compared to the saturation fluence. The change in the stored fluence, of course, is exactly equal in magnitude, under the assumption of no losses, to the amount of fluence extracted from the medium which goes into the signal pulse. Thus,

$$\Delta J_{stor} = J_{stor,i} - J_{stor,i-1} = -(J_i - J_{i-1}) = -\Delta J_i,$$  \hfill (25)

so we can rewrite the gain update as

$$g_i = g_{i-1} - \frac{\Delta J_i}{J_{sat}}.$$  \hfill (26)

Now if we recall equation 23, we can relate the small signal gain to the small gain cross section as

$$g = \sigma_g N$$  \hfill (27)
and thus relate the small signal to the inverted fraction by

\[ g_i = g_{i-1} - \frac{\Delta J_i}{J_{sat}} \]

\[ g_i = g_{i-1} - \frac{\Delta J_i}{hcl(\sigma_{em} + \sigma_{abs})} \]

\[ N(\beta_i(\sigma_{em} + \sigma_{abs}) - \sigma_{abs}) = N(\beta_{i-1}(\sigma_{em} + \sigma_{abs}) - \sigma_{abs}) - \frac{\Delta J_i}{hcl(\sigma_{em} + \sigma_{abs})} \]  

(28)

and thus solving for \( \beta_i \) get

\[ \beta_i = \beta_{i-1} - \frac{\Delta J_i}{hclN(\sigma_{em} + \sigma_{abs})}. \]  

(29)

Now in the chromatic case, the change in the extracted energy from the medium is precisely the sum of the difference in the current fluence, adjusted for losses, and the previous fluence over all wavelengths. Thus we can arrive at the final equation for updating the inversion factor as

\[ \beta_i = \beta_{i-1} - \frac{\int \lambda \left( \frac{J_i(\lambda)}{T(\lambda)} - J_{i-1}(\lambda) \right) d\lambda}{hclN}. \]  

(30)

The final two equations needed for the simulation for updating fluence and inversion fraction are

\[ J_i(\lambda) = J_{sat}(\lambda)T(\lambda) \ln \left\{ 1 + \left[ e^{\frac{J_{i-1}(\lambda)}{J_{sat}(\lambda)}} - 1 \right] G_0(\lambda) \right\} \]  

(31)

and

\[ \beta_i = \beta_{i-1} - \frac{\int \lambda \left( \frac{J_i(\lambda)}{T(\lambda)} - J_{i-1}(\lambda) \right) d\lambda}{hclN}. \]  

(32)

Then while running the simulation, the input fluence is sliced such that the flattop signal assumption under which the fluence solution was derived is still held. Each slice of the fluence can independently be updated based on the inversion factor. Then the slices can be summed to arrive at a final output fluence for one pass through the amplifier. This procedure repeats for each pass through the amplifier.

The signal amplification stage is then followed by a pumping stage to reintroduce energy into the gain medium. The pumping process follows the exact same equations and only requires one pass.

Therefore, the simulation first requires the proper amplifier parameters (see table 1) and can account for intricacies in the gain medium such as etalons, rotated crystal orientation, and upper-state lifetime of the medium during the pumping process by adjusting amplifier parameters and mixing cross-section data. Second, the model requires an input fluence shape for the seed laser.
This essentially amounts to an adjustment of the spectral amplitude profile of the laser. For the simulation, it is assumed that no additional substantial phase is added by the amplifier, especially compared to the phase added by any pulse shaping or upconversion. However, a constant added phase can be easily incorporated into the model if there is a known phase after amplification.

| Abbreviation | Parameter | Units          |
|--------------|-----------|----------------|
| φ            | photon density | photons/m²     |
| N₁           | number density ground state electrons |                    |
| N₂, nₑ       | number density excited state electrons |                    |
| σ            | resonance absorption cross section |                |
| β            | inverted fraction |               |
| G(λ)         | spectral single pass gain |                |
| g(λ)         | spectral gain | m⁻¹              |
| σₚ(λ)        | spectral gain cross section | m²              |
| σₑₘ         | spectral emission cross section | m²              |
| σₑₐₛ        | spectral absorption cross section | m²              |
| J(λ)         | spectral fluence | J/m²          |
| N            | dopant ion density | m⁻³            |
| l            | length of gain medium | m               |
| T(λ)         | single pass transmission |               |
| c            | speed of light | m/s             |
| h            | Planck’s constant | Js               |

Table 1

3.1 Upconversion

The upconversion method is unique compared to the pulse shaping and the amplification as there are many methods for nonlinear upconversion and each one is significantly different from the others. As our work is following the example of the photo-injector laser at SLAC’s LCLS-II, we also limit our model discussion to their setup. The photo-injector laser uses dispersion controlled nonlinear shaping (DCNS), which uses noncolinear sum frequency generation to combine two highly dispersed pulses followed by second harmonic generation for the final upconversion to UV [17]. Figure 3 shows the schematic setup of the mixing scheme where, for this particular use case, a copy of the IR pulse is made and the now, two IR pulses are shaped with equal and opposite chirp.

Modeling the setup requires solving the wave equation [47] for each field as shown below

$$\nabla^2 E_n(\omega) - \frac{\epsilon^{(1)}(\omega_n)}{c^2} \frac{\partial^2 E_n(\omega)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 P_n^{NL}(\omega)}{\partial t^2}$$

(33)

where \(E_n\) is the particular electric field, \(P_n^{NL}\) is the associated nonlinear polarization, \(\omega_n\) is the associated frequency, \(\epsilon^{(1)}\) is the scalar dielectric constant, \(\epsilon_0\) is the permittivity of free space, and \(c\) is the speed of light. Then for sum-frequency generation the nonlinear polarization for the output frequency is
given by

\[ P(\omega_3) = 4\epsilon_0 d_{\text{eff}} E(\omega_1)E(\omega_2) \]  \hspace{1cm} (34)

where \( d_{\text{eff}} \) represents the \( \chi \) nonlinearity tensor or contracted matrix.

The electric fields are treated in the frequency domain as \( E(\omega) = A(\omega)e^{i\phi(\omega)} \) where the spectral phase \( (\phi(\omega)) \) is treated with a Taylor expansion around \( \omega_0 \) and accounts for the added chirp to electrics fields one and two, \( E_1(\omega) \) and \( E_2(\omega) \). Then in the time domain the fields are \( E_i(z,t) = E_i e^{-i\omega_i t} + \text{c.c.} \) where \( E_i = A_i e^{ik_i z} \). Subbing in the forms of the electric fields we get

\[ P_3 = 4\epsilon_0 d_{\text{eff}} A_1 A_2 e^{i(k_1+k_2)z} = p_3 e^{i(k_1+k_2)z}. \]  \hspace{1cm} (35)

This result can now be used in the wave equation to get

\[
\begin{align*}
\left[ \frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} - k_3^2 A_3 + \frac{\epsilon^{(1)}(\omega_3)\omega_3^2 A_3}{c^2} \right] e^{i(k_3 z - \omega_3 t)} + \text{c.c} \\
= -\frac{4d_{\text{eff}}\omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2)z - \omega_3 t} + \text{c.c.}
\end{align*}
\]  \hspace{1cm} (36)

The equation can be simplified since \( k_3^2 = \epsilon^{(1)}(\omega_3)\omega_3^2/c^2 \) and because we can drop the complex conjugates and still maintain the equality. Furthermore, we can use the slowly varying wave approximation as well and repeat the process for each field [17, 47]

\[
\begin{align*}
\frac{dA_1}{dz} &= 2id_{\text{eff}}\omega_1^2 \frac{1}{k_1 c^2} A_3 A_2^* e^{-i\Delta k z} \\
\frac{dA_2}{dz} &= 2id_{\text{eff}}\omega_2^2 \frac{1}{k_2 c^2} A_3 A_1^* e^{-i\Delta k z} \\
\frac{dA_3}{dz} &= 2id_{\text{eff}}\omega_3^2 \frac{1}{k_3 c^2} A_1 A_2 e^{i\Delta k z}.
\end{align*}
\]  \hspace{1cm} (37) (38) (39)

where \( \Delta k = k_1 + k_2 - k_3 \).
The coefficients in equations 37–39 can be reformulated in terms of index of refraction resulting in

\[
\frac{2i d_{\text{eff}} \omega^2}{k_i c^2} = \frac{2i d_{\text{eff}} 2\pi}{\lambda_i n_i(\lambda)}
\]

(40)

where \(\lambda_i\) is the central wavelength of the corresponding field and \(n_i(\lambda)\) is the wavelength-dependent index of refraction for that field.

The index of refractions can then be calculated using the chosen Sellmeier equations. Here \(A_1\) and \(A_2\) are along the ordinary axis for the crystal and for a BBO crystal have index of refraction

\[
n_o = \sqrt{A_o + \frac{B_o}{\lambda^2} + C_o + D_o \lambda^2}
\]

(41)

where \(A_o\), \(B_o\), \(C_o\), and \(D_o\) are chosen for the particular crystal of interest [48]. For \(A_3\), the output is at an angle between the ordinary and extraordinary axes so can first calculate the extraordindary axis index of refraction using

\[
n_e = \sqrt{A_e + \frac{B_e}{\lambda^2} + C_e + D_e \lambda^2}
\]

(42)

where \(A_e\), \(B_e\), \(C_e\), and \(D_e\) are chosen for the particular crystal of interest [48], and then can mix the two axes using

\[
n_{e\theta} = \frac{1}{\sqrt{\cos^2 \theta + \sin^2 \theta}}
\]

(43)

where \(\theta\) is the angle.

Finally, these equations are solved using a symmetrized split-step fourier method to handle propagation in the crystal and then a fourth-order Runge-Kutta algorithm solves the coupled nonlinear equations [17].

4 Results

As seen in section 2, the S2E model has far ranging applications. With the current state of the model, there are four primary use cases presented here: 1) reverse engineering CPA systems; 2) CPA system optimization; 3) inverse design for CPA systems; 4) integration of CPA and NLO systems.

4.1 Reverse Engineering

Matching the behavior of the amplifier model to the physical device is essential for accurate simulation. As seen in section 3, the amplifier model depends on many parameters. Typically, a physical CPA device manufacturer does not disclose all of the required parameters. However, one may have some initial guess or intuition on parameters that work physically. Figure 4(a) shows the
input spectral amplitude to the amplifier model and experiment (in green), the experimental output spectral amplitude (in orange), and a non-optimized simulation spectral amplitude output (in blue). This non-optimized spectrum is modeled from an arbitrarily selected set of parameters that seem like plausible physical values. Specifically these parameters are 10 mm for the crystal length, \(7 \times 10^{24} \text{ 1/m}^3\) dopant ion density, input seed laser radius of 400 \(\mu\text{m}\). However, we see that the simulated result does not exhibit sufficient gain-narrowing and is not located at the correct central wavelength. Furthermore, the energy build-up in the multipass amplifier (shown in figure 4 (b)) exhibits relatively low gain.

Fig. 4

Clearly, one must tune the model parameters in order to obtain a set of parameters that yield simulated behavior similar to experimental results. There are several approaches for tuning these parameters, but here we choose to scan over a set of parameters and compare the output spectral amplitude to the experimental amplifier output spectral amplitude using normalized root mean-squared error. We then looked for minimum error that also yielded a physical result. We also set additional requirements on the output from the simulation, including a minimum energy threshold and a saturation condition for the amplification process. Here we focused our parameter scans on crystal length, mode radius for the pump, dopant ion density, and the mixing of the three crystal axes. The crystal length and dopant ion density were then combined into one parameter as their product is the only quantity relevant in the equations (see section 3).

Figure 5 shows the heat maps for the error minimization for portions of these scans. Specifically, each subfigure shows a different crystal axis tuning for the same scan over radius and combined parameter. The figures show several regions of lowest error in the dark regions. These represent potential solutions. The structure of the error heat maps suggest stability regions of operation for particular combinations of parameters. On each plot, the minimum error solution is highlighted with a blue box and the maximum error solution is
Fig. 5

highlighted with a red box. Figures 5 (b) and (c) show the output spectral amplitude for the minimum error solution out of all the error plots shown and the maximum error solution, respectively.

Fig. 6

We can then look at this minimized error solution on this subset of scans in the context of the experimental results. Figure 6 (a) shows again the input to the amplifier model and physical device (in green), the experimental output (in orange), and the minimum error solution from this scan (in blue). We can see this optimized solution much more closely matches the desired experimental spectrum than the non-optimized, arbitrarily selected parameters in figure 4 (a). Also, we can see the energy build up as a function of the number of passes in 6 (b). Here the energy does not saturate, which matches a known condition
of this particular amplifier. Furthermore, the final output energy demonstrates a build-up rather than a decay, a necessary requirement for an amplifier.

After running the error minimization routine to achieve a result that matches the experimental result, one might consider ways to optimize the experimental system or ways to improve on a follow-up design of the amplifier to better match the requirements in the lab. This leads to using the S2E model for optimization of CPA systems.

4.2 CPA Optimization

The spectral output characteristics of a CPA play an essential role in the CPA system as a whole. Mainly this constitutes the output spectral amplitude bandwidth, energy, shape, and central wavelength. If one is designing a CPA from the ground-up, then there are many degrees of freedom in the design. One must choose the material, the crystal length, the crystal orientation, the number of round-trip passes, the presence of any etalons or notches, etc. Simulation can help narrow down these choices in order to achieve the desired amplifier. Even once the amplifier is built with the particular physical elements in place, there are still a number of parameters that can be used to optimize the operation of the amplifier. For instance, the pump power and number of round-trip passes in the amplifier can be tuned to alter the final output characteristics. It is useful to explore this parameter space in simulation in order expedite such tuning in lab.

To demonstrate the optimization for ground-up design and for post-design tuning, we take the spectral amplitude output result from section 4.1 as a given amplifier and attempt to alter the output bandwidth, central wavelength, and energy of the final result. Specifically, we investigate number of roundtrip passes, pump power, and the presence of various spectral notches in the amplifier. The first two parameters can be tuned before or after amplifier construction. The spectral notch would normally be chosen during amplifier design and not tuned after construction unless one has an open design for the amplifier.

First, we adjust the number of passes through the amplifier without changing anything else. Figures 7 (a)–(c) correspond to the same amplifier simulation parameters as resulted from the reverse engineering in section 4.1 except the number of passes go up to 66. Figure 7 (a) shows the spectral amplitude output of every tenth roundtrip pass. Here we can see the peaks are clearly growing and the profile is narrowing. Inset figure 7 (b) shows the output spectral amplitude of the 10th, 20th, and 30th pass in the amplifier. Figure 7 (c) then shows the corresponding energy buildup for each pass. Here the pulses reach saturation around 66 roundtrips. After saturation the energy transfer from amplification medium to laser pulse becomes less efficient and even reverses. Furthermore, as the number of passes increases, the effect of gain-narrowing becomes more pronounced. This could be useful in cases that want to use a more narrow spectral output for resonance. Conversely, this narrowing can be detrimental to cases requiring ultrafast applications with extremely short pulses in time.
Then we can increase the pump power. Figures 7 (d)–(f) repeat the results for figures 7 (a)–(c) except the pump power is increased by 67%. Here we see the same buildup and pulse characteristics except saturation occurs earlier at around 56 roundtrip passes, as expected.

Therefore, in tuning number of passes, there is a balance in bandwidth and energy, but if one wants to keep the pulse characteristics relatively constant but boost energy, then increasing pump power is an appropriate option.

Of course, the more degrees of freedom in the search, the more combinations one can achieve. For instance, if one could change the absorption and emission cross-sections for the amplification medium, then the central wavelength location and gain-narrowing effect could be altered in a way to precisely achieve a specific output bandwidth and energy. This leads into the capabilities of using this S2E for inverse design.
4.3 Inverse Design

The question becomes whether one can now select desired amplifier output characteristics and use similar error minimization and optimization routines to instead tune parameters focused around material properties to design new amplification mediums or find new potential uses for existing materials. While exploring this fully is outside the realm of this paper, we show a simple example that begins how one would approach this use-case for the S2E model. Assume we want an output spectrum centered close to 1025 nm with a broader spectrum than the reverse engineered result with a BW close to 10 nm and non-saturated energy build up after 30 passes in the amplifier, as shown in figure 8.

![Graph showing absorption and emission cross sections for Yb:KGW](image)

Fig. 8

The task then becomes to find absorption and emission cross sections for the crystal that would yield this result. Such a search is complicated by the fact that the other parameters in the model also affect the output characteristics; nevertheless, we show a simplified search here. We can explore perturbations on commonly used amplification mediums.

Figure 9 shows examples of potential modifications on emission and absorption cross sections for one crystal axis for Yb:KGW, essentially defining a new material, and the corresponding amplifier spectral amplitude simulation outputs. For instance in figure 9 (a) and (b), broadening the emission peak and boosting its cross-section yields an output with a dominant peak and smaller secondary peak at a slightly higher wavelength because of the broader emission spectrum around 1025 nm. Figure 9 (c) and (d) has the emission cross section boosted less than in (a) and the drop-off in the emission spectra peak around 1025 nm leads to more pronounced hole in the amplifier simulation output in (d). We also see here a significant reduction in energy compared to (b) since the emission cross section is significantly smaller. Similarly the output is not saturated after 30 passes. Finally 9 (e) shows the spectra that leads to the desired spectrum from figure 8. To achieve this broader bandwidth, mixed crystal axes as well; however, doing so leads to the lower final output...
energy. While further exploration of this search is outside the purview of this work, the next steps would be to perform iterative backpropagations or other optimization searches to find the required cross sections and then to work with material scientists to ensure the results are physically realizable with the correct dopants and base substrates.

4.4 CPA and NLO Integration

Beyond the CPA alone, start-to-end simulations incorporating nonlinear devices and NLO systems play a crucial role in real world applications. However, important subtleties are often overlooked in simulating cascaded processes, especially the amplifier’s role in altering the spectrum, the phase
transfer through the nonlinear optical devices, and thus the resulting time-domain intensity profile. Here, we demonstrate the importance of properly simulating each module as a part of the total integrated model. As our example usecase for an integrated system focuses on the photo-injector laser for LCLS-II, our modeled components include a pulse shaper, amplifier, and upconversion using SFG and SHG. However, applications beyond the photo-injector laser benefit from the accurate and efficient modeling of these components. We explore here the combination of pulse shaper and amplifier and of pulse shaper, amplifier, and upconversion.

Combining a pulse shaper and CPA gives significant more flexibility for altering the output characteristics from the amplifier pulse since, as discussed in section 3, the pulse shaper performs spectral phase and amplitude shaping. Phase shaping allows for control over temporal pulse width and chirp. This provides a means for altering the time-domain output of the amplifier without affecting the spectral intensity’s characteristics, and thus retaining spectral bandwidth. By using combinations of second, third, and fourth order phase, one can achieve particular time-domain intensity profiles; however, after amplification, these profiles are often quite different.

Figure 10 shows the intensity and phase for the pulse shaper output (blue) and amplifier output (yellow) in the time-domain (top row, (a)–(d)) and frequency domain (bottom row, (e)–(f)) for various combinations of SOD, TOD, and 4OD. For example, in wakefield accelerators, a highly sought after electron-bunch charge distribution, which can in theory be achieved with a triangular shaped laser pulse driving the production of the electrons [49]. Figures 10 (a) and (e) show the time-domain triangular pulse and corresponding frequency domain phase. However, we see that after amplification, the time-domain intensity changes drastically away from the desired triangle pulse at the output of the pulse shaper. Other applications such as pump lasers for OPCPA systems [50] and photo-injector lasers [28] benefit from flat-top pulses for higher efficiency energy transfer for amplification and reduced electron beam emittance, respectively. Figures 10 (b) and (f) show a non-optimized square pulse with oscillations in the time-domain and frequency domain. After amplification, there is actually a reduction in the pulse duration and a reduction in the
oscillations. Figures 10 (c), (d), (g), and (h) are relevant in applications requiring an initial higher intensity pulse followed by an additional pulse, as might be the case in electron beam applications attempting to stimulate microbunching. Once again, though, we can see how including the amplification process can reduce or alter the nature of these trailing pulses.

Fig. 11

Beyond just phase shaping, amplitude shaping by carving a hole in the spectral amplitude opens more possibilities for shaping. For instance if we want to achieve a more triangular-shaped pulse after amplification, then a different set of dispersion parameters in conjunction with amplitude shaping can be used. Figures 11 (a), (d), (e), and (h) show the time and frequency domains for the pulse shaper and amplifier outputs that yield amplifier outputs resembling triangular-shaped pulses. For these we can see in the frequency domain the main difference came down to the position and size of the hole being carved in the spectral amplitude as the phase was kept approximately the same. Furthermore, we can see that the pulse shaper output in both cases does not intuitively lead to the amplifier output shape. For flat-top pulse shapes or flat-top shapes with oscillations, figures 11 (b), (f), and (g) show possible selections for hole positions, widths, and depths to achieve a relatively flat-top pulse after amplification. Yet, once again, we can clearly see how the time domain output from the pulse shaper is not the same as the time domain output from the amplifier. This important when using a simulation to inform system design or parameter choices as these types of differences can either set requirements for different equipment choices or explain unexpected laboratory observations.

Many applications require higher harmonics with specific or tunable pulse shapes, which can be produced via NLO systems proceeding amplification [51]. Such a setup mimics the setup shown in figure 1 with mode-locked oscillator, pulse shaper, amplifier, and nonlinear upconversion. As mentioned previously, since we are using the photo-injector laser for LCLS-II as an example, then the nonlinear upconversion involves noncolinear SFG followed by a spectral 1 nm bandpass filter and SHG to result in a final UV output.

Figure 3 shows the time domain intensity profiles and phases for the pulse shaper (blue), amplifier (yellow), and final SHG (green) outputs. The standard
output with no phase shaping is shown in (a). For photo-injector lasers, there is interest in being able to control asymmetries in the output, achieving flat-top or modulated flat-top pulses, or achieving Gaussian upconverted signals. Furthermore, it is clear that the phase does not directly transfer in many nonlinear schemes. Figure 3 (b) shows the ability to control asymmetries in the lobes of the SHG output signal using added dispersion in the pulse shaper. Figure 3 (c) shows an output approaching a flat-top pulse with a central peak, and figure 3 (d) shows an output resembling a broadened Gaussian. For (c) and (d), holes were carved in the spectrum using spectral amplitude shaping at the pulse shaper, corresponding to figure 11 (b) and (c), respectively.

5 Conclusion

The advent of the CPA has had a dramatic impact on society with high-power laser systems affecting everything from medical procedures and diagnoses to national defense and security. In order continue driving progress in the field and to prepare for an age focused around applying machine learning requiring large data sets to CPA and NLO systems, accurate and efficient modeling is crucial. Our S2E model can help fill this simulation gap by providing a modular and expandable simulator that can be used for everything from reverse engineering hardware components, to optimization of current system designs, to inverse design of materials for amplifiers, to full-scale simulation of optical systems for data generation.

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