

Numerical and experimental study of an ultrabroadband modeless Ce:LiCAF ultraviolet laser: Multiwavelength dynamics and power amplification

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Abstract. *This study investigates the generation and power amplification of an ultrabroadband modeless cerium-doped lithium calcium aluminum fluoride (Ce:LiCAF) ultraviolet (UV) laser system operating in an ASE-dominated regime using combined numerical and experimental approaches. Pumped at 266 nm, the Ce:LiCAF system produces a modeless UV spectrum from 275 to 330 nm with a dominant peak at ≈ 288.5 nm (full width at half maximum, FWHM ≈ 16 nm) and a weaker band near 310 nm. We develop a multiwavelength propagation model in which the spectrum is represented by discrete wavelength channels with wavelength-dependent emission and absorption cross-sections, and we include pump absorption, gain saturation and an effective wavelength-dependent loss that accounts for excited-state absorption (ESA) and reabsorption. The modeless seed spectrum is described by a semi-empirical model based on the intrinsic emission cross-section $\sigma_e(\lambda)$, and the four-pass amplifier is simulated using a spectrally resolved Frantz–Nodvik formalism. Experimentally, a four-pass Ce:LiCAF amplifier is implemented, and spectra and pulse energies are measured after the modeless seed branch and after each amplifier pass at a fixed 266 nm pumping energy. The amplified source delivers up to 1 mJ per pulse, with the FWHM of the 288.5 nm peak reduced from 16 nm to 7 nm, indicating moderate gain narrowing while preserving the two-band spectrum. Numerical predictions agree with the measurements (deviations $<10\%$ in output energy and <1 nm in peak wavelength), showing that this modeless Ce:LiCAF system is a promising source for broadband UV absorption spectroscopy and trace-gas detection.*

Keywords: Ce:LiCAF ultraviolet laser; multi-pass amplifier; multiwavelength modeling; ASE-dominated modeless UV laser.

Classification numbers: 42.55.Rz; 42.60.Da; 42.60.By; 42.72.Bj.

1. Introduction

Broadband ultraviolet (UV) sources are important for applications such as differential optical absorption spectroscopy (DOAS) and trace-gas monitoring, where wide spectral coverage and sufficient pulse energy are required to resolve weak absorption features [1, 2]. Among available UV gain media, cerium-doped fluoride crystals, in particular Ce:LiCAF, are attractive because the allowed $5d-4f$ transition of Ce^{3+} provides a broad emission band in the 275–330 nm region together with a short fluorescence lifetime and high quantum efficiency at room temperature [1–5]. Ce:LiCAF lasers have been demonstrated in a variety of configurations, including continuous-wave and pulsed operation, narrow-linewidth and broadband regimes, total-internal-reflection cavities and systems optimized for spectroscopy [5–8]. These works show that Ce:LiCAF can deliver high-brightness UV radiation in compact sources suitable for laboratory and field measurements.

A promising route to exploit the full gain bandwidth of Ce:LiCAF is to operate in a “modeless” or amplified spontaneous emission (ASE)-dominated regime [1], where feedback is weak or highly multimode and the output is not constrained by a discrete set of longitudinal modes. In this manuscript, the term “modeless (ASE-dominated) Ce:LiCAF UV laser” refers to broadband UV emission generated under weak and highly multimode feedback, where the output is initiated by spontaneous emission and shaped by stimulated amplification in the gain medium, without selection of discrete longitudinal cavity modes. Such ASE-dominated modeless configurations naturally support very broad spectra but are more difficult to model because gain and loss are strongly wavelength dependent (emission/absorption/ESA and reabsorption), so single-wavelength models cannot capture the resulting spectral reshaping and gain narrowing.

In previous work we demonstrated an ultrawide-band modeless Ce:LiCAF UV source for SO_2 detection using DOAS [1, 2], focusing on the design and characterization of the modeless seed branch. Here we extend this approach by investigating both the modeless source and its power amplification in a four-pass Ce:LiCAF amplifier using a multiwavelength numerical model validated by experiment. The model represents the spectrum by discrete wavelength channels with wavelength-dependent gain and loss and includes pump absorption and gain saturation, with the modeless seed spectrum described by a semi-empirical approach based on the emission cross-section $\sigma_e(\lambda)$. Experimentally, we measure spectra and pulse energies after the modeless seed branch and after each amplifier pass at a representative pump level. The main outcome is a validated description of the energy scaling and spectral evolution of a power-scaled modeless Ce:LiCAF UV source that maintains a broad two-band spectrum around 288.5 and ~310 nm and is suitable for broadband UV absorption spectroscopy and trace-gas detection. For clarity, we note that a spectrally resolved (chromatic) Frantz–Nodvik propagation framework was also used in our previous four-pass Ce:LiCAF amplifier study [9]. In the present work, however, the modeling is developed and used for a different regime: a broadband modeless/ASE-type seed covering a wide UV band and exhibiting a two-band structure (around 288.5 nm and ~310 nm). Therefore, the numerical treatment is configured to resolve the full-band spectral evolution and capture gain

sharing/competition between the two emission bands during amplification, which is not required in the narrowband resonator-seeded regime.

2. Multiwavelength theoretical model

We describe the broadband gain dynamics of the modeless Ce:LiCAF UV source and its four-pass amplification using a one-dimensional, multiwavelength model. The 275–330 nm emission band is represented by discrete wavelength bins, and the spectral fluence in each bin is propagated through (i) a modeless ASE-dominated seed branch and (ii) a spectrally resolved Frantz–Nodvik amplifier. The spectroscopic parameters for Ce:LiCAF are taken from Refs. [3, 4], while the concept of modeless operation follows Ref. [1]. Unlike the narrowband resonator-seeded configuration considered in Ref. [9], the present model is intended for a broadband two-band modeless seed; accordingly, it explicitly tracks the spectral evolution over the full band and the gain competition between the ~288.5 nm and ~310 nm features during propagation.

2.1. Scope and assumptions

The gain medium is a Ce:LiCAF crystal of length L , pumped at $\lambda_p = 266$ nm and operated with nanosecond pulses, so a rate-equation and fluence description is appropriate. The model is one-dimensional along the crystal axis. We assume that pump and signal share an effective mode area A_{eff} (corresponding to a Gaussian mode with radius w_s) and describe incomplete overlap with the doped region through confinement factors Γ_p and Γ_s . The UV emission band is discretized into wavelength bins centered at λ_i with a step of $\Delta\lambda = 0.1$ nm. For each bin, we use π -polarized emission and absorption cross-sections $\sigma_e(\lambda_i)$, $\sigma_a(\lambda_i)$ and a normalized spontaneous-emission spectrum $B(\lambda_i)$ derived from the digitized Ce:LiCAF π -polarized emission cross-section spectrum reported in Refs. [3, 4]. $B(\lambda_i)$ is normalized such that $\sum_i B(\lambda_i)\Delta\lambda = 1$. Optional ESA, reabsorption, and scattering are included in an effective distributed loss coefficient $\alpha(\lambda_i)$, and the total Ce^{3+} concentration is N_{tot} with fluorescence lifetime τ_f . Unless otherwise stated, the simulations use the same crystal length, Ce^{3+} concentration, pump pulse energy and approximate spot sizes as in the experiment of Sec. 3.

2.2. Modeless seed branch and seed spectrum

In the experimental modeless configuration, the Ce:LiCAF crystal is pumped at 266 nm and emits broadband UV radiation dominated by ASE, which is collected and redirected by a concave mirror [1]. A full time-dependent ASE model would require solving coupled rate and transport equations for the forward and backward radiation fields [10, 11]. Here we adopt a simpler semi-empirical description that preserves the essential spectral features of the seed while keeping the numerical model tractable.

Conceptual ASE description

Conceptually, the Ce^{3+} ions are modeled as an effective two-level system with ground- and excited-state populations $N_1(z, t)$ and $N_2(z, t)$ (with $N_{\text{tot}} = N_1 + N_2$). The upper-state population obeys a rate equation of the form

$$\frac{dN_2(z,t)}{dt} = \eta_p \sigma_{ap}(\lambda_p) \Phi_p(z,t) N_1(z,t) - \frac{N_2(z,t)}{\tau_f} - \Gamma_s \sum_i \frac{I_i(z,t)}{h\nu_i} [\sigma_e(\lambda_i) N_2(z,t) + \sigma_a(\lambda_i) N_1(z,t)], \quad (1)$$

where $\sigma_{ap}(\lambda_p)$ is the pump absorption cross-section at 266 nm, $\Phi_p(z,t)$ is the pump photon flux, η_p is an effective pump quantum efficiency including branching and spatial overlap, $I_i(z,t)$ is the spectral radiant power density at λ_i , and $\nu_i = \frac{c}{\lambda_i}$.

For a given inversion profile $N_2(z)$, the small-signal gain coefficient at wavelength λ_i is

$$g_i(z) = \Gamma_s [\sigma_e(\lambda_i) N_2(z) - \sigma_a(\lambda_i) N_1(z)], \quad (2)$$

and the spectral intensity in a one-dimensional ASE model would obey

$$\frac{\partial I_i(z,t)}{\partial z} = g_i(z) I_i(z,t) + S_{sp,i}(z,t) - \alpha(\lambda_i) I_i(z,t), \quad (3)$$

with $S_{sp,i}(z,t)$ a spontaneous-emission source term proportional to $N_2(z,t) B(\lambda_i)$. In a pure ASE configuration (i.e., neglecting weak feedback in the present seed branch), the boundary conditions are $I_i(0,t) = 0$ and $I_i(L,t)$ taken as the output. The corresponding seed spectral fluence at the modeless seed output (i.e., at the amplifier input in our measurements) would be

$$F_{seed}(\lambda_i) = \int I_i(L,t) dt. \quad (4)$$

Practical construction of the seed spectrum from $\sigma_e(\lambda)$

Instead of solving Eqs. (1)–(3) explicitly for all λ_i - which would require detailed knowledge of the feedback geometry, collection solid angle and spatial pump profile in the modeless cavity - we adopt a semi-empirical approach in which the spectral shape of the seed is approximated by the π -polarized emission cross-section $\sigma_e(\lambda)$ of Ce:LiCAF. A practical advantage of using $\sigma_e(\lambda)$ as the seed lineshape, rather than the experimentally recorded seed spectrum, is that $\sigma_e(\lambda)$ is an intrinsic material quantity obtained from carefully calibrated spectroscopic measurements [3,4], whereas the measured seed spectrum may retain residual uncertainties from the detection chain (spectrometer throughput, detector spectral sensitivity, fiber-coupling efficiency), even after instrumental-response correction, particularly in the wing regions. The comparison between the $\sigma_e(\lambda_i)$ -based seed and the measured seed spectrum then serves as an independent validation of the model input (see Sec. 4.1, Fig. 2). Under these conditions, we construct a normalized seed shape

$$S_{seed}(\lambda_i) = \frac{\sigma_e(\lambda_i)}{\sum_j \sigma_e(\lambda_j) \Delta\lambda}, \quad (5)$$

and assign a total seed energy E_{seed} based on the pump energy at 266 nm, the effective absorbed fraction, the ASE efficiency and the seed collection efficiency,

$$E_{seed} = E_{ps} f_{abs} \eta_{ASE} \Gamma_{seed}, \quad (6)$$

where E_{ps} is the pump pulse energy directed to the modeless configuration, f_{abs} is the fraction absorbed in the crystal, η_{ASE} is an effective ASE conversion efficiency, and Γ_{seed} accounts for spatial overlap and collection by the concave mirror. The corresponding total seed fluence is

$$F_{tot} = \frac{E_{seed}}{A_{eff}}, \quad (7)$$

and the spectral fluence at the amplifier entrance becomes

$$F_{\text{seed}}(\lambda_i) = F_{\text{tot}} S_{\text{seed}}(\lambda_i). \quad (8)$$

Equations (5)–(8) provide a semi-empirical seed model whose spectral shape is governed by the intrinsic emission cross-section $\sigma_e(\lambda)$ and whose total energy is anchored to the absorbed 266 nm pump energy through effective efficiency factors. The detailed ASE build-up, feedback geometry and collection optics are treated implicitly via the lumped efficiency factors η_{ASE} and Γ_{seed} (and the absorbed-pump fraction f_{abs}). Because $\sigma_e(\lambda)$ is used as the model input, the seed spectrum entering the amplifier simulation is determined entirely by material parameters and is independent of the instrumental response of the experimental detection chain. The validity of this approximation is confirmed by direct comparison with the independently measured seed spectrum (Section 4.1, Fig. 2): The FWHM values agree within ~ 0.7 nm (16.4 nm for the $\sigma_e(\lambda)$ -based seed versus 15.7 nm measured) and both spectra exhibit the same two-band structure around 288.5 nm and ~ 310 nm. The remaining discrepancies in the long-wavelength wing are discussed in more detail in Sec. 4.1.

2.3. Spectral Frantz–Nodvik amplification

For the four-pass amplifier we treat the seed as a nanosecond pulse characterized by its spectral fluence $F_{\text{seed}}(\lambda_i)$ at the amplifier entrance. In the present amplifier model, we propagate the injected seed fluence using the spectral Frantz–Nodvik formalism. Self-generated ASE inside the amplifier is not explicitly included and is expected to contribute mainly as a weak broadband background in the present non-resonant four-pass geometry. For a given pass n and wavelength bin λ_i , a pumped crystal with initial inversion $N_{2,0}^{(n)}$ amplifies the input fluence $F_{\text{in}}^{(n)}(\lambda_i)$ according to the Frantz–Nodvik law applied independently at each λ_i [9, 12]:

$$F_{\text{out}}^{(n)}(\lambda_i) = F_{\text{sat}}(\lambda_i) \ln \left[1 + \exp(G_0(\lambda_i)) \left(\exp \left[\frac{F_{\text{in}}^{(n)}(\lambda_i)}{F_{\text{sat}}(\lambda_i)} \right] - 1 \right) \right], \quad (9)$$

with saturation fluence and small-signal gain exponent

$$F_{\text{sat}}(\lambda_i) = \frac{\frac{hc}{\lambda_i}}{\sigma_e(\lambda_i)}, \quad (10)$$

$$G_0(\lambda_i) = \Gamma_s \sigma_e(\lambda_i) N_{2,0}^{(n)} L - \alpha(\lambda_i) L.$$

Here $F_{\text{sat}}(\lambda_i)$ is the intrinsic saturation fluence of the gain medium. The corresponding saturation energy for the UV mode is $E_{\text{sat}}(\lambda_i) = A_{\text{eff}} F_{\text{sat}}(\lambda_i) / \Gamma_s$, i.e. only the fraction Γ_s of the mode overlapping the doped region contributes to gain saturation [2–4, 8]. Reabsorption and other wavelength-dependent attenuation are included in the effective loss term $\alpha(\lambda)$; therefore $G_0(\lambda)$ is written using $\sigma_e(\lambda)$ and $\alpha(\lambda)$ as an effective net gain exponent.

Passive losses from the amplifier optics in pass n are included through a wavelength-dependent one-pass transmission $T^{(n)}(\lambda_i) \in (0, 1]$,

$$\tilde{F}_{\text{out}}^{(n)}(\lambda_i) = T^{(n)}(\lambda_i) F_{\text{out}}^{(n)}(\lambda_i). \quad (11)$$

Photon extraction during pass n reduces the inversion. To account for the wavelength-dependent photon extraction, we implement a wavelength-resolved depletion bookkeeping based

on the extracted photon number in each λ_i bin:

$$\Delta N_2^{(n)}(\lambda_i) \propto \frac{F_{\text{out}}^{(n)}(\lambda_i) - F_{\text{in}}^{(n)}(\lambda_i)}{\frac{hc}{\lambda_i}}. \quad (12)$$

This procedure yields a wavelength-resolved estimate of depletion contributions, while the physical inversion is shared across the band and is reduced by the total extracted photons. In practice, this factor acts as a regularization to prevent numerical over-depletion in a single pass while preserving the overall saturation behaviour constrained by the measured energy scaling. Because the four passes occur within a single pump pulse, we neglect any recovery of the inversion between passes ($\Delta t \ll \tau_f$), so that the inversion is only reduced by gain depletion and not replenished by the pump.

The four-pass amplifier is simulated by iterating Eq. (9) for $n = 1, \dots, 4$, starting from $F_{\text{in}}^{(1)}(\lambda_i) = F_{\text{seed}}(\lambda_i)$, and using $F_{\text{in}}^{(n+1)}(\lambda_i) = T^{(n)}(\lambda_i) F_{\text{out}}^{(n)}(\lambda_i)$ for the subsequent passes. This multiwavelength Frantz–Nodvik description captures the wavelength-dependent gain saturation, gain narrowing around the main emission peak near 288.5 nm, and the evolution of the weaker long-wavelength band around 310 nm.

2.4. Integral outputs and link to measurements

From the amplified spectral fluence we derive the quantities directly compared with experiment. For pass n , the total spectral fluence is

$$F_{\text{tot}}^{(n)} = \sum_i F_{\text{out}}^{(n)}(\lambda_i) \Delta \lambda, \quad (13)$$

and the corresponding output pulse energy and average power for repetition rate f_{rep} are $E^{(n)} = A_{\text{eff}} F_{\text{tot}}^{(n)}$, $P_{\text{avg}}^{(n)} = f_{\text{rep}} E^{(n)}$. The normalized output spectrum used in the comparison plots is defined as

$$S^{(n)}(\lambda_i) = \frac{F_{\text{out}}^{(n)}(\lambda_i)}{\max_j F_{\text{out}}^{(n)}(\lambda_j)}, \quad (14)$$

from which we extract the FWHM of the main peak around 288.5 nm and track the evolution of the weaker band near 310 nm as a function of pass number. By adjusting a small set of effective parameters (overlap factors, internal loss slope, ASE efficiency and depletion strength) and from the experimental setup in Section 3, the model reproduces the measured evolution of the output energy, gain narrowing (FWHM from ≈ 16 nm to ≈ 7 nm) and preservation of the two-band spectral structure across the four-pass amplifier, as discussed in Sec. 4.

3. Experiment

Figure 1 shows the experimental layout of the ultrabroadband modeless Ce:LiCAF UV system and the four-pass amplifier. The fourth harmonic (266 nm) output of a Q-switched Nd:YAG laser (LS-2137U-N, Lotis TII, pulse duration 7 ns; repetition rate 10 Hz) is routed to the experiment by three steering mirrors M_1 – M_3 , which are dielectric UV mirrors with high reflectivity at 266 nm at 45° incidence. After M_2 the beam is split by a 50/50 beam splitter (BS) into two branches: one pumps the modeless seed branch (ASE-dominated Ce:LiCAF emission), and the other pumps the four-pass amplifier, as indicated in Fig. 1. The four-pass amplifier layout follows

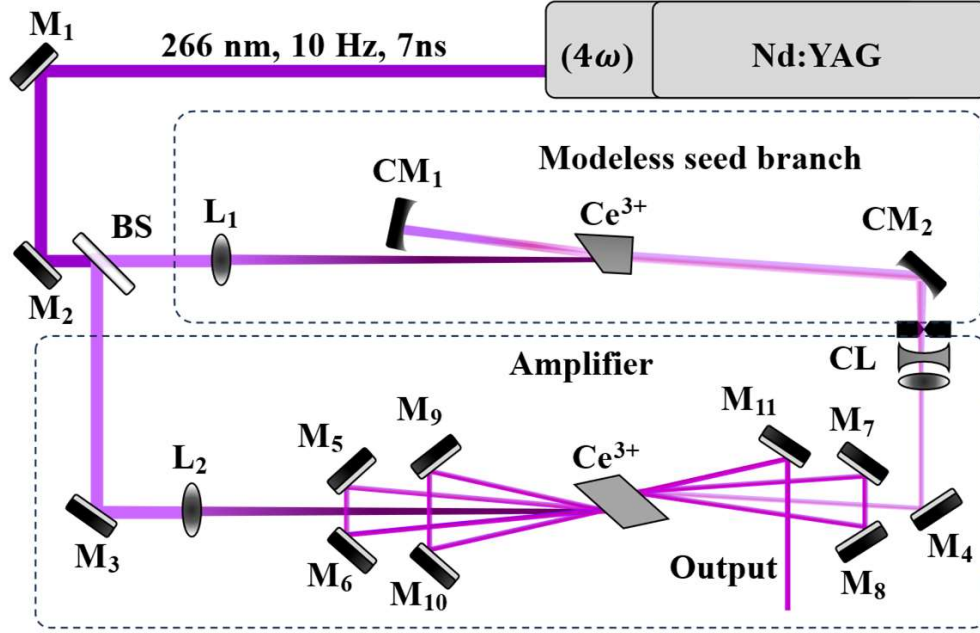


Fig. 1. Experimental setup of the broadband modeless Ce:LiCAF UV source and four-pass Ce:LiCAF amplifier pumped by the 266 nm fourth harmonic of a Q-switched neodymium-doped yttrium aluminum garnet (Nd:YAG) laser. Only the main mirrors and lenses are shown for clarity. The four-pass amplifier stage is adapted from our previous implementation (Ref. [9]), while the modeless seed branch is redesigned here for broadband ASE-dominated (modeless) operation.

our previously reported design [9], and is reused here as a stable amplification platform; the seed-generation branch is redesigned for the modeless broadband source. The pump pulse energies delivered to the Ce:LiCAF crystals are set to $E_{po} \approx 10$ mJ (modeless seed branch) and $E_{pa} \approx 11$ mJ (amplifier branch), measured after the BS and the delivery optics.

3.1. Modeless Ce:LiCAF UV source

In the modeless seed branch, the 266 nm beam transmitted by the beam splitter is focused into a Ce:LiCAF crystal by a fused-silica lens L_1 . In the experiment, we use a 10 mm long Ce:LiCAF crystal doped with 1 mol% Ce³⁺. The crystal is cut in a trapezoidal geometry, with one end face polished at the Brewster angle for π -polarized radiation near 290 nm and the opposite face polished perpendicular to the beam; both faces are left uncoated. The pump beam is weakly focused on an approximately Gaussian spot with a radius of about $w_p \approx 1$ mm at the crystal. With a pump pulse energy of 10 mJ in this branch, the corresponding pump fluence is ~ 0.32 J.cm⁻². The crystal is cut and oriented so that the laser polarization is π -polarized ($E \parallel c$ -axis), matching the emission and absorption cross-sections used in the simulations (subsection 2.1). In this branch, CM₁ and CM₂ are UV-enhanced aluminum mirrors optimized for the 250–450 nm range, providing >90% average reflectance over 250–450 nm according to the manufacturer's specifications (Thorlabs), while the lenses (L_1 , CL) are UV fused-silica elements with antireflection coatings at

245–400 nm, providing low loss for both the pump and the broadband UV emission. The seed branch is operated deliberately in an ASE-dominated, modeless regime: the Ce:LiCAF crystal is pumped close to normal incidence, and a concave mirror CM₁ placed near the pumped face collects a fraction of the spontaneous emission/ ASE and redirects it through the crystal. The feedback is weak and highly multimode, so no discrete longitudinal modes are selected [10]. The emerging broadband UV beam (“modeless seed” in Fig. 1) is folded by CM₂ and collimated by a lens CL before entering the amplifier branch.

3.2. Four-pass Ce:LiCAF amplifier

The second pump branch passes through a shaping lens L₂ (Fig. 1) before entering the amplifier crystal, which is another Ce:LiCAF crystal with similar length and doping, also oriented for π polarization, cut as a parallelogram rod with both end faces polished at the Brewster angle for π -polarized radiation near 290 nm. The pump spot radius w_p and the seed mode radius w_s in the amplifier crystal are adjusted to be comparable, consistent with the effective overlap factor Γ used in the multiwavelength model. The shaping lens L₂ is used to condition the 266 nm pump beam and to obtain a stable pump spot in the amplifier crystal. In the one-dimensional model, transverse pump/seed profile details are represented by an effective overlap factor Γ (and effective mode area), so residual pump non-uniformity contributes mainly to the uncertainty of Γ discussed in Sec. 4.2.

In the experiments reported here, the 266 nm pump pulse energy in the amplifier branch is set to about $E_{pa} \approx 11$ mJ, corresponding to the operating point used for the numerical–experimental comparison. At this setting, the modeless seed delivers a seed energy of ~ 0.2 mJ and the amplifier outputs energies up to ≈ 1 mJ per pulse after the fourth pass.

A four-pass geometry is implemented using UV-enhanced aluminum mirrors M₄–M₁₁. After the first pass through the amplifier crystal, the UV beam is imaged back onto the crystal for the second, third and fourth passes, with the propagation distances chosen so that the beam size in the crystal remains nearly constant. The cumulative transmission of the mirrors, beam splitter and crystal coatings define the effective path efficiencies $T^{(n)}(\lambda_i)$ used in Section 2.3. After the fourth pass, the amplified beam is directed towards the diagnostics as the system output.

3.3. Diagnostics

A small fraction of the UV beam after the modeless seed branch and after each amplifier pass is sampled by thin fused-silica plates and sent to a fiber-coupled UV spectrometer (AvaSpec-HSC1024x58TEC-EVO, 200–385 nm range, ~ 0.4 nm resolution). The recorded spectra are corrected for the instrumental response and used to determine the spectral shape and FWHM of the main 288.5 nm peak and the evolution of the weaker ~ 310 nm band.

Pulse energies at the output of the modeless seed branch and after each pass are measured with a Coherent LabMax-TOP energy meter equipped with an LM-2UV detector head. At the chosen 266 nm pump energy (kept fixed for both the modeless seed branch and the amplifier), spectra and pulse energies are averaged over many shots to reduce pulse-to-pulse fluctuations. These measurements provide the experimental seed and amplified energies, spectral widths and peak positions at this operating point, which are compared with the predictions of the multiwavelength model in Sec. 4.

4. Results and discussion

4.1. Seed spectrum of the modeless Ce:LiCAF UV source

Figure 2 shows the normalized spectrum of the modeless Ce:LiCAF UV source at the operating point used in this work (solid black curve, “Exp. seed”) together with the seed spectrum used in the simulations (dashed red curve, “Sim. seed”). The measured modeless output extends over roughly 275–330 nm, with a dominant peak at $\lambda \approx 288.5$ nm and a weaker secondary band near 310 nm on an asymmetric broadband pedestal. The full width at half maximum (FWHM) of the main peak is approximately 15.7 nm, confirming that the modeless seed branch provides an ultrabroadband UV seed suitable for broadband absorption spectroscopy [1, 3, 4].

In the numerical model, the seed spectrum is constructed from the intrinsic π -polarized emission cross-section $\sigma_e(\lambda)$ of Ce:LiCAF as described in the semi-empirical model of Section 2.2. The tabulated $\sigma_e(\lambda)$ is interpolated on the simulation wavelength grid, normalized to unit peak, and then scaled in energy (before normalization for plotting) so that the integrated seed energy matches the experimentally measured value at the amplifier entrance. The resulting numerical seed spectrum (Sim. seed) reproduces well the measured spectral profile: the peak position at 288.5 nm, the FWHM (16.4 nm simulated versus 15.7 nm measured), and the presence of the weaker long-wavelength band around 310 nm are all captured within the experimental and modeling uncertainties. This close agreement confirms that the $\sigma_e(\lambda)$ -based semi-empirical seed model of Section 2.2 provides an adequate description of the modeless seed spectrum and that the use of the intrinsic emission cross-section free from instrumental distortions is a reliable basis for the amplifier simulations.

Some discrepancies remain in the long-wavelength wing beyond ~ 305 nm, where the measured spectrum shows a slightly different curvature and relative amplitude of the ~ 310 nm band. These differences are expected because the semi-empirical seed model is based on the intrinsic $\sigma_e(\lambda)$ lineshape and does not explicitly include wavelength-dependent reabsorption, ESA, spatial pump inhomogeneity, or the exact optical geometry and feedback conditions in the modeless seed branch. As a result, the simulated seed spectrum should be regarded as an effective approximation to the actual modeless output. When used as the input to the amplifier simulation, the remaining mismatch mainly appears as a small uncertainty in the effective overlap and loss parameters, without affecting the main amplification trends. Overall, the $\sigma_e(\lambda)$ -based seed model provides a physically consistent and experimentally anchored input for the multiwavelength Frantz–Nodvik amplifier simulations discussed in Sections 4.2 and 4.3.

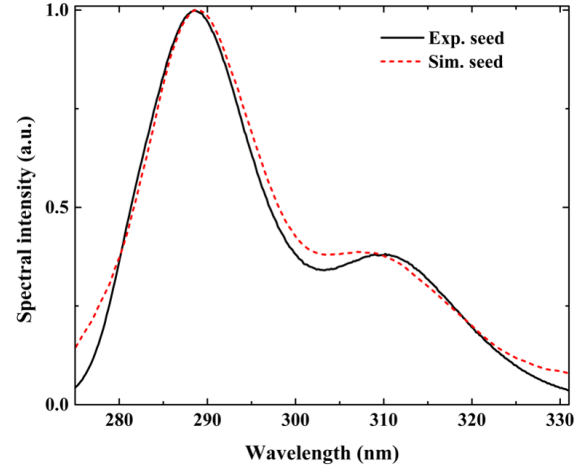


Fig. 2. Normalized seed spectra of the modeless Ce:LiCAF UV source. The solid black curve (Exp. seed) shows the measured modeless output at the amplifier input, and the dashed red curve (Sim. seed) shows the $\sigma_e(\lambda)$ -based seed used in the simulations.

4.2. Energy scaling in the four-pass Ce:LiCAF amplifier

The energy scaling of the modeless Ce:LiCAF UV source in the four-pass amplifier is summarized in Fig. 3. At the chosen 266 nm pump energy, the modeless seed branch delivers a seed pulse energy of about 0.2 mJ at the amplifier input ($n = 0$). As the seed propagates through the amplifier, the measured pulse energy increases monotonically and reaches ≈ 1 mJ after the fourth pass, i.e. an overall energy scaling by a factor of about five. The per-pass gain $G_{\text{pass}} = \frac{E_{\text{out}}^{(n)}}{E_{\text{in}}^{(n)}}$ decreases from the value of the order of three on the first pass to slightly above unity on the fourth pass, reflecting progressive gain saturation and finite internal losses. The amplified energy reported here (~ 1 mJ after four passes from a ~ 0.2 mJ seed) reflects the broadband modeless seeding regime, in which the spectral fluence at any given wavelength is lower than in a narrowband seed of the same total energy, resulting in weaker gain saturation and reduced energy extraction. In addition, the ~ 310 nm band shares the same population inversion and can, to a limited extent, redistribute a fraction of the extracted energy across the spectrum rather than concentrating it near 288.5 nm.

In the simulations, the total pulse energy after each pass is obtained by integrating the multiwavelength Frantz–Nodvik output fluence over wavelength, using the same crystal length, pump energy and approximate mode size as in the experiment. The effective overlap factors, wavelength-dependent loss $\alpha(\lambda)$ and spectral depletion strength are adjusted once so that the simulated seed and fourth-pass energies match the experimental values within their uncertainties. With these parameters fixed, the model predicts the energy evolution across the four passes without further tuning. As shown in Fig. 3, the simulated pulse energies and per-pass gains follow the measured ones for all passes, with deviations typically within about 5–10 %.

The remaining discrepancies can be attributed to uncertainties in the effective mode area, the exact pump and seed beam radii in the crystal, and the cumulative transmission of the optical components, none of which are known with high precision. Small misalignments or beam clipping in the experimental amplifier arm, which are not included in the one-dimensional model, may further reduce the measured gain compared with the idealized case. Nevertheless, the good agreement in Fig. 3 indicates that the chosen inversion, loss and overlap parameters provide a consistent description of the amplifier and that the multiwavelength Frantz–Nodvik model correctly reproduces the energy scaling of the broadband Ce:LiCAF UV source at this operating point.

The present simulations and measurements are validated at a representative operating point (10 mJ in the modeless seed branch and 11 mJ in the amplifier branch). Since the net gain exponent $G_0(\lambda)$ in Eq. (10) is set by the pump-generated inversion, reducing the pump energy can lead to negligible amplification, whereas increasing it drives stronger saturation and more

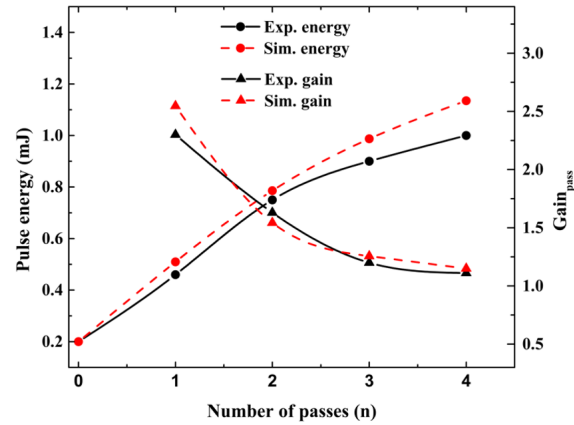


Fig. 3. Measured and simulated pulse energy (left axis) and per-pass gain $\text{Gain}_{\text{pass}}$ (right axis) of the four-pass Ce:LiCAF amplifier as a function of the number of passes n ($n = 0$ corresponds to the seed).

pronounced gain narrowing. A systematic mapping of output energy and bandwidth versus pump energy, including identification of the minimum pump level for appreciable amplification and the onset of strong saturation, will be addressed in future work.

4.3. Spectral evolution and gain narrowing in the amplifier

The spectral evolution of the broadband Ce:LiCAF UV pulse in the four-pass amplifier is shown in Fig. 4, where normalized experimental spectra (solid black) are compared with simulated spectra (dashed red) for passes 1–4. After each pass the spectrum retains the characteristic two-band structure with a dominant band around 288.5 nm and a weaker band near 310 nm but undergoes gradual gain narrowing and a slight shift of the main peak. The measured FWHM of the main band decreases from ~ 15.7 nm for the modeless seed to ≈ 14 nm after the first pass, ≈ 11 nm after the second pass, ≈ 9 nm after the third pass and ≈ 7 nm after the fourth pass. This behaviour is typical of broadband amplifiers with spectrally peaked gain: wavelengths near the maximum of $\sigma_e(\lambda)$ experience higher gain and saturate first, while the wings are comparatively suppressed. The multiwavelength Frantz–Nodvik model reproduces this gain-narrowing trend; the simulated FWHM values differ from the measurements by at most ~ 1 nm, confirming that the wavelength-dependent emission cross-section and loss $\alpha(\lambda)$ used in the model provide a realistic gain profile.

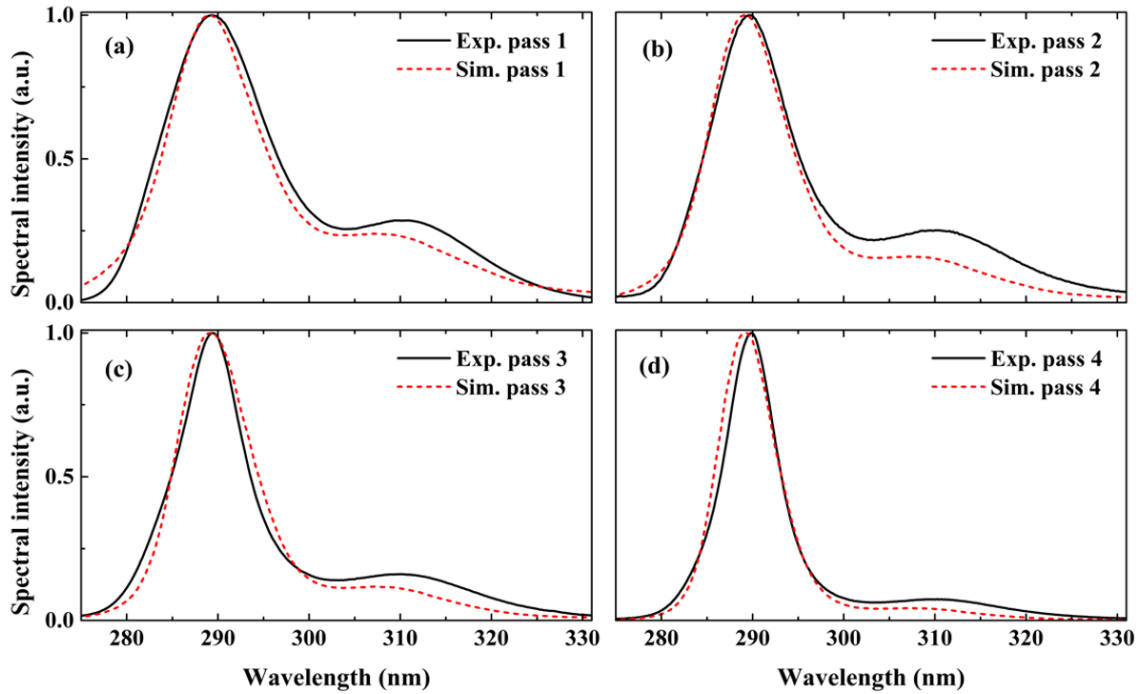


Fig. 4. Normalized experimental (solid black) and simulated (dashed red) spectra of the Ce:LiCAF UV pulse after (a) pass 1, (b) pass 2, (c) pass 3 and (d) pass 4 in the four-pass amplifier. Spectra are normalized to the peak of the main band to highlight gain narrowing and the evolution of the weaker ~ 310 nm wing.

A small redshift of the spectral peak is also observed. The peak wavelength moves from $\lambda_{\text{peak}} \approx 288.5$ nm for the modeless seed to about 289.8 nm after the fourth pass. This can be attributed to wavelength-dependent losses in the amplifier beam path: the reflectivity of the aluminum-coated mirrors increases gradually from 275 to 300 nm and continues to rise toward a maximum around 320 nm for the mirrors used in our setup, so wavelengths slightly longer than 288.5 nm experience slightly lower loss than those on the short-wavelength side. In the simulations, this behaviour is represented by a gently sloping loss term $\alpha(\lambda)$; the resulting peak shift is of the same order as in the experiment.

The weaker band around 310 nm is progressively reduced in relative amplitude as the pulse passes through the amplifier, but it does not vanish even after four passes, indicating that the net gain at these longer wavelengths remains positive at the chosen pump level. The model reproduces the overall trend of suppression of the ~ 310 nm band, with slightly stronger attenuation than observed experimentally, which is consistent with the conservative choice of $\alpha(\lambda)$ and spectral inversion depletion. The persistence of the 310 nm band confirms that the power-scaled source maintains a genuinely broadband two-band spectrum, an important feature for broadband UV absorption spectroscopy and DOAS applications.

4.4. Pulse duration

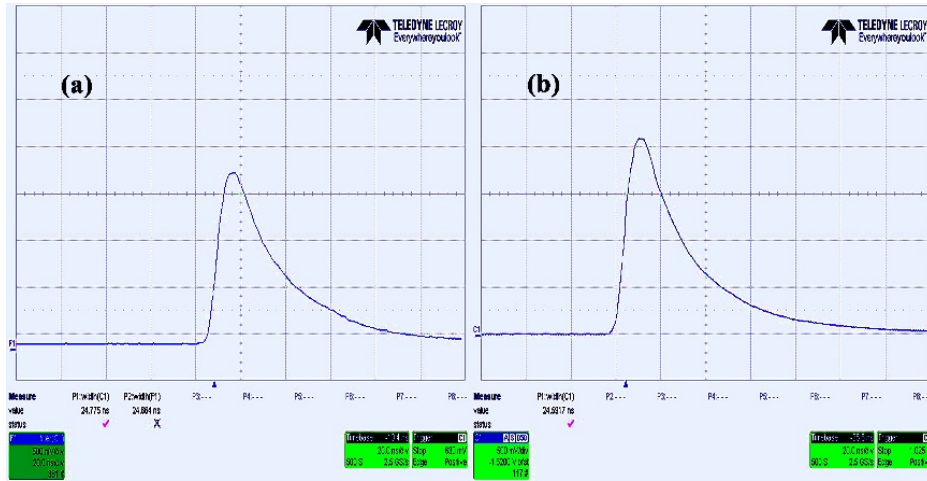


Fig. 5. Measured temporal profiles of the modeless UV pulse (a) at the output of the modeless seed branch and (b) after the fourth pass of the amplifier. Both traces are recorded with a fast photodetector and oscilloscope at the operating pump energy, showing similar pulse shapes with FWHM durations of about 25 ns.

To characterize the temporal behaviour of the broadband Ce:LiCAF UV source, we measured the pulse duration of both the modeless seed output and the amplified pulses using a fast photodiode (Hamamatsu S9055) with a response time of about 233 ps, coupled to a 5 GHz digital oscilloscope (WaveMaster 8600A, LeCroy). Fig. 5 shows typical temporal profiles of the UV pulse at the output of the modeless seed branch and after the fourth pass of the amplifier. In both cases, the pulse exhibits a FWHM duration of about 25 ns. This value is significantly longer

than the 7 ns duration of the 266 nm pump pulse and is comparable to the intrinsic fluorescence lifetime of Ce:LiCAF, consistent with an ASE-dominated modeless emission regime rather than a short mode-locked or Q-switched pulse.

Within the accuracy of our measurements, the four-pass amplification stage does not noticeably change the pulse duration: the amplified pulses in Fig. 5 remain close to 25 ns while their energy and spectrum evolve as described in Subsections 4.2 and 4.3. Although dynamic gain depletion can lead to pulse reshaping (often pulse shortening) in high-gain amplifiers, such an effect is not observed here. This is consistent with our operating regime, where the broadband modeless seed energy is low (~ 0.2 mJ) and the extraction is distributed over four passes, resulting in weak dynamic saturation within the ~ 25 ns envelope. This supports the use of an energy/fluence-based, quasi-steady-state description in the multiwavelength amplifier model of Sec. 2, where the four passes are treated as occurring within a single excitation window and temporal reshaping effects are neglected compared with spectral gain narrowing and wavelength-dependent losses.

5. Conclusion

We have investigated power amplification of an ultrabroadband modeless (ASE-dominated) Ce:LiCAF UV laser system using a combined numerical–experimental approach. The modeless seed delivers a broadband UV spectrum spanning ~ 275 to 330 nm with a dominant peak at 288.5 nm and a weaker band near 310 nm. A semi-empirical seed model based on the intrinsic π -polarized emission cross-section $\sigma_e(\lambda)$, validated against the measured seed spectrum at the amplifier input, provides a consistent and instrument-independent spectral input for the simulations.

The spectrally resolved multiwavelength Frantz–Nodvik model, including wavelength-dependent gain, loss and spectral depletion, successfully describes the pass-to-pass energy and spectral evolution at the investigated operating point. Experimentally, the pulse energy increases from ~ 0.2 mJ to ~ 1 mJ after four passes, while the main band narrows from ~ 16 nm to ~ 7 nm and the peak shifts slightly from 288.5 nm to ~ 289.8 nm. Simulated energies and spectra agree with the measurements typically within 5–10%, and the amplified output retains the characteristic two-band structure around 288.5 and ~ 310 nm.

The measured pulse duration of ~ 25 ns, comparable to the Ce:LiCAF fluorescence lifetime and essentially unchanged by amplification, supports operation in an ASE-dominated modeless regime and justifies a fluence-based, quasi-steady-state description. Future work will address the pump-level dependence of the amplification dynamics, including identification of the minimum pump level for appreciable amplification and the onset of strong saturation.

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