Development of Future All-Solid-State, Ultraviolet, Terawatt Laser System using Ce:LiCAF as a Gain Medium

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Chirped-pulse amplification (CPA) in the ultraviolet region is demonstrated by using a broadband Ce:LiCAF laser medium. For the terawatt (TW) CPA system, the 25-fs, 290-nm pulses are successfully generated by the hollow-fiber pulse compression. Additionally, a coaxially pumped, large-aperture ultraviolet power-amplifier is demonstrated to have 98-mJ output with stable pump source using LB4 crystals. These elements will be the key devices for terawatt-class ultraviolet Chirped-pulse amplification laser systems.

High-peak-power, femtosecond, ultraviolet (UV) laser have recently attracted a large amount of interest for material processing applications [1] in addition to the traditional scientific applications [2]. There have been three approaches for UV high-peak power lasers. One is the nonlinear frequency conversion of the Ti:sapphire amplifier system [3]. For frequency conversion, the UV output energy is strongly limited by the available nonlinear crystal size. The other approach is KrF excimer amplifiers [4]. However, KrF excimer amplifiers have a fundamental limitation in tunability and achievable pulse duration because of their gain bandwidth. The final approach is the chirped pulse amplification (CPA) laser system using UV gain medium. Ce:LiCaAlF6 (C:LiCAF), which is the tunable solid-state UV laser medium with sufficient broad bandwidth (280-320 nm) and a high saturation fluence exceeding 100 mJ/cm² from its emission-cross section [5]. Previously, we demonstrated the first

Fig. 1 (a) Broadened bandwidth and (b) transmittance of the pulses from the fiber filled with the gases: neon, argon, krypton, and xenon at different gas pressures. Argon is the best choice considering the broadened bandwidth and transmittance.
UV CPA using a broad-band Ce:LiCAF crystal as the gain medium [3]. The peak power of the amplified and compressed pulse (τ ~ 115 fs) at 290 nm reached to 30 GW. For further peak-power scaling up to terawatt (TW) level, one can try to compress the pulse width and improve the output pulse energy.

In this letter, we report the generation of intense 15-µJ, 25-fs pulses at 290 nm by using a hollow fiber filled with high-pressure argon gas and a coaxially pumped large-aperture power-amplifier module with stable pump source using LB4 crystals. Additionally, the detailed design parameters for further energy scaling are successfully evaluated toward TW UV Ce:LiCAF laser system.

Since the Ce:LiCAF crystal has a broad tunability from 281 nm to 315 nm, it shows the possibility of the generation and amplification of 3-fs pulse [6-9]. Such ultraviolet pulse is required as the seed pulse of TW-class Ce:LiCAF laser system. To date, a powerful pulse-compression technique, which is based on self-phase-modulation (SPM) [10] in a hollow fiber filled with noble gases, has been demonstrated [11-15]. It is reasonable to select a very short pulse with a broad-spectrum width for the seed pulses of the TW-class UV CPA system. The femtosecond seed pulses at 290 nm with 1-kHz repetition rate were provided by frequency tripling of the output from the Ti:sapphire regenerative amplifier. The durations of 290-nm femtosecond pulses were evaluated by a third-order autocorrelator using the visible fluorescence of the XeF C-A transition [16]. A photographic lens with sufficient resolution enlarged the fluorescence image. The duration of the seed pulse at 290 nm was measured to be 210 fs. The seed pulses were guided and focused into the hollow fiber. The fused silica hollow fiber is 22 cm in length and 250 µm in diameter. The fiber was kept straight in the chamber filled with noble gases. We tested four kinds of gases: neon, argon, krypton, and xenon. Neon gas had a larger transmittance, but its broadened bandwidth was small as shown in Fig. 1.

For krypton and xenon gases, the broadened bandwidths became larger while their transmittances became smaller when the gas pressures were increased. So the choice of argon gas as the nonlinear medium is a good compromise between a moderate ionization threshold and moderate nonlinearity. The output pulse energy

![Figure 2](image)

Fig. 2 (a) Spectra of the seed pulses before and after the fiber filled with argon gas at 7-atm pressure. (b) Third-order autocorrelation trace obtained when the distance between the two prisms was set at 30 cm. The pulse was compressed down to 25 fs duration assuming a sech pulse shape with a time and bandwidth product of 0.37.
amounts to approximately 24 µJ, implying a throughput of 34%. The spectra of the seed pulses before and after the fiber are shown in Fig. 2(a), the spectral bandwidth is broadened to 4.2 nm due to SPM. The spectra-broadened pulse was guided to the dispersion compensating, double-pass, and Brewster quartz prism pair. Figure 2(b) shows the autocorrelation trace obtained when separation between the two prisms was set at 30 cm. The pulse was compressed down to 25 fs duration, which gave a time and bandwidth product of 0.37. The compressed pulse had pulse energy of 15 µJ [17]. These ultrashort pulses will be very suitable to be used as the seed pulses of TW-class Ce:LiCAF laser system.

![Fig. 3. CZ-grown LiCAF boule with an 11-cm diameter. The growth direction is along the c-axis.](image)

The other devises for TW UV all-solid-state lasers are the pumping source for Ce:LiCAF crystal and the scalable power-amplifier-module design that is capable of handling high pulse energy in UV region. The fourth harmonics of Nd:YAG lasers is an ideal pumping source because of the absorption spectra of Ce:LiCAF crystal. We have succeeded to generate the total forth harmonics output energy (266nm) of 430 mJ with the total conversion efficiency of 30.5%, using three Li₂BaO₇ (LB₄) crystals [18]. Compared with BaB₂O₄ (BBO) and CsLiB₆O₁₀ (CLBO), LB₄ crystal has advantages in the aspects of low absorption in the UV region, excellent mechanical properties, easy fabrication, and high resistance to humidity. Moreover, 30-% conversion efficiency in total was maintained for more than 15 hours without any adjustment. By such performance of pump source, we can redesign a power-amplifier module for a TW-class UV laser system [19]. The Ce:LiCAF crystal used for first UV CPA was rather small (1 x 1 x 1 cm³). We successfully grew a 7-cm diameter Ce:LiCAF crystal [20], and are still endeavoring to grow larger LiCAF crystals for deep-ultraviolet lithographic applications at F₂ laser wavelengths shown in Fig. 3 [21].

The experimental setup of a coaxially-pum ped large-aperture Ce:LiCAF double-pass power-amplifier module is illustrated in Fig. 4(a). A large-sized Brewster-plate gain module (1 x 2 x 2 cm³) with a 3.2-cm⁻¹ absorption coefficient at 266-nm and a 0.2-cm⁻¹ absorption coefficient at 290-nm was cut from a 5-cm diameter Ce:LiCAF boule. For the characterization of this module, a conventional Ce:LiCAF master oscillator and pre-amplifier delivered 15-mJ seed pulses at 290 nm. To pump the power-amplifier module, we prepared four beams of 266-nm pulses from three 10-Hz, Q-switched Nd:YAG lasers with a total excitation energy of 0.38-J using LB₄ crystals. From the output-energy
dependence on different input energy as shown in Fig. 4(b), the gain saturation was clearly observed. The small-signal gain and the saturation fluence were evaluated to be 39 times and 77 mJ/cm² on the assumption of Frantz-Nodvik relation. The highest output energy reached to 98 mJ for 13 mJ seed pulse with the extraction efficiency of 25%. The amplified spontaneous emission (ASE) was below the detection limit. For the output energy scaling design with variety of pump and input energy, the small signal gain coefficient dependence for different pump density was evaluated as shown in Fig. 4(c).

Based on this small-signal gain coefficient the extraction efficiency and output energy for different pump diameter was estimated for as shown in Fig. 5(a). Considering the damage threshold of the gain medium for 266-nm pump pulse and that of the high-reflection coating for 290-nm pulses, the energy limitation factor will be the damage of
turning mirror for 290-nm as shown in Fig. 4(a), and thus the pulse width of seed pulses has to be stretched over 10ps. From these results, the system as shown in Fig. 5(b) is designed to generate the TW-class UV pulses.

![Diagram](image)

**Fig. 5.** (a) Calculated output fluence and extraction efficiency for different beam diameter with the fixed pump energy. The damage thresholds of the coating for shorter pulse duration are also calculated according to a $\tau^{1/2}$ dependence of the damage threshold. The output fluence for shorter pulse durations is limited by the damage threshold of mirrors. (b) Future design of the TW-class UV laser system.

In conclusion, we have successfully demonstrated the generation of 15-µJ, 25-fs pulses at 290 nm and a coaxially pumped large-aperture Ce:LiCAF power-amplifier module to have 98-mJ output with 25% extraction efficiency. These performances of the seed pulse and the gain module are adequate for TW-class UV CPA laser systems. Such solid-state laser development will open up a new category of high-power lasers for real world applications as an alternative to Ti:sapphire CPA laser systems.

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