Mode-locked deep ultraviolet Ce:LiCAF laser

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We report mode-locked operation of a synchronously pumped Ce:LiCAF oscillator. The laser operated in the deep UV with output radiation centered at 291 nm and a pulse duration of 6 ps. The maximum output power measured was 52 mW, with 13% slope efficiency. The Ce:LiCAF crystal has a gain bandwidth capable of supporting few-femtosecond pulses, and so our results demonstrate the potential to form a new class of ultrafast lasers operating directly at deep UV wavelengths. © 2009 Optical Society of America

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Ultrashort UV laser sources are important tools for a wide range of applications across science, including probing of extremely fast physical and chemical processes [1], investigating the relaxation of charge carriers in semiconductors [2], or controlling ultrafast chemical processes at the atomic level [3,4]. The lack of robust, simple, and reliable ultrafast laser sources that directly generate deep UV radiation has inhibited the study of ultrafast processes requiring tunable deep UV ultrashort pulses.

To address this concern, a variety of laser systems has been proposed. Most are based on amplified Ti:sapphire systems and rely on a range of $\chi^{(2)}$ and $\chi^{(3)}$ processes to generate short pulses in the deep UV [5–9]. For example, Graf *et al.* generated 4 fs pulses by harmonic generation in a gas jet [10]; they produced 1.4 μJ pulses at around 275 nm but required a regeneratively amplified 6 fs, 0.25 mJ Ti:sapphire laser, achieving an efficiency of 0.6% and operating at 1 kHz.

There are clear advantages to generating ultrafast radiation directly at deep UV wavelengths. Cerium-based lasers are well-established efficient sources for tunable output in the UV spectral region from 280 nm up to 345 nm [11] but to date have provided only gain-switched pulses with durations of hundreds of picoseconds or longer [12]. The particular crystal Ce:LiCAF is conveniently pumped at 266 nm by the fourth harmonic of a Nd:YAG laser and offers a gain bandwidth of more than 30 nm with a peak gain at 290 nm. This bandwidth is in principle capable of supporting pulses as short as 3 fs, with the single-cycle limit at 290 nm in the attosecond domain.

The short laser upper level lifetime of Ce:LiCAF of just 25 ns means that high pump power densities are required to achieve cw and mode-locked operation [13]. Mode-locked picosecond Nd:YAG lasers can be frequency quadrupled to generate quasi-cw radiation at 266 nm with high efficiency, fulfilling this requirement and providing a route to mode locking of a synchronously pumped cerium laser. In this work, we report what we believe to be the first demonstration of such a mode-locked Ce:LiCAF oscillator, generating UV pulses as short as 6 ps with a slope efficiency up to 13%.

The experimental setup of our laser system is shown in Fig. 1. The diode-pumped cw mode-locked Nd:YAG laser (Photonic Industries PS300-1064) produced 28 ps pulses at a repetition rate of 78 MHz. The average 1064 nm output power was 22 W. The IR beam was first chopped to avoid thermal effects in the doubling and quadrupling stages, generating 100 μs duration bursts of pulses every 1.3 ms. This radiation was frequency doubled by noncritically phase-matched second-harmonic generation in a 3.5-cm-long lithium triborate (LBO) crystal heated to 144°C. Stable output powers of 13 W were obtained (note that all powers are quoted as the average burst power while the chopper was open; the average power over the full chopper cycle was 1/13 of that power). The 532 nm radiation was then line focused into a 4-mm-long β-barium borate (BBO) crystal, generating powers of 2 W at 266 nm with a pulse duration of 23 ps.

The Ce:LiCAF oscillator operated with a three-mirror astigmatically compensated cavity as shown in Fig. 1. The 3.5% cerium-doped rod, held in a copper mount at room temperature, was cut at Brewster's angle with its c axis perpendicular to the propagation direction and in the horizontal plane. The pump was horizontally (π) polarized to minimize excited state absorption in the crystal [14]. The 1.25-mm-long crystal absorbed 70% of the pump radiation. The pump beam was matched to the cavity mode by lens L1 (5 cm focal length) through the dichroic mir-

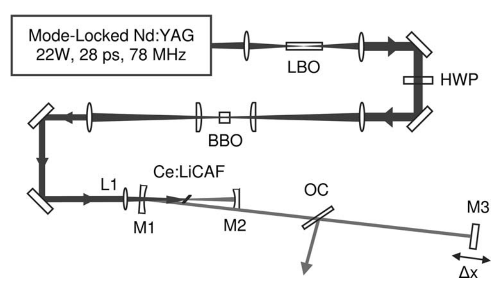

Fig. 1. Experimental setup: L1, spherical lens focal length $f=5$ cm; M1, M2, concave spherical mirrors with ROC (radius of curvature)=10 and 5 cm, respectively; M3, plane mirror; OC, output coupler; HWP, half-wave plate at 532 nm.

ror M1, producing an approximately 16 μm diameter beam waist. The transmission at 266 nm of the dichroic mirror and the focusing optics was 68%, and so the maximum absorbed pump power was 990 mW.

The three cavity mirrors had low transmission ($<0.03\%$), and so a 6-mm-thick UV-grade silica plate placed in the cavity at close to Brewster's angle was used as a variable output coupler. The cavity lengths of the pump and cerium laser were matched for synchronous mode locking. Each time the chopper opened a stable mode-locked pulse train was produced after approximately 4 μs build up time. A maximum output of 52 mW was obtained, with an output coupling of 3% inferred by measurement of the coupler angle. All reflections from the plate were included in the power measurement; this power could be easily produced in a single output beam with a suitable transmissive cavity mirror. In this configuration, the lasing threshold was 580 mW of the absorbed pump power, and the slope efficiency was 13%. The variation in the output power versus cavity length is depicted in Fig. 2, where output powers of over 50 mW were obtained for translations of $\sim$100 μm around the length for perfect synchronization. Moving M3 more than 0.5 mm away from synchronization, the cavity produced a constant output of 25 mW.

The measurement of pulse duration at deep UV wavelengths for subnanojoule pulse energy is challenging. Since second-harmonic generation cannot be employed for these short wavelengths, most UV autocorrelation methods rely on $\chi^{(3)}$ nonlinearities, requiring pulse energies of hundreds of nanojoules per pulse [15]. Increased sensitivity can be achieved using a $\chi^{(2)}$ cross-correlation technique, but this requires an auxiliary short pulse to either sum- or difference-frequency mix with the UV pulse. Since the 1064 nm pump pulses were too long to be used effectively for this purpose, we instead used a femtosecond Ti:sapphire laser running at 800 nm, which we difference-frequency mixed with the 291 nm Ce:LiCAF pulses to generate a signal at 457 nm. Since the Ti:sapphire and cerium lasers were not synchronized, we employed an asynchronous sampling method [16]. The lasers were adjusted to have a small offset between their repetition rates, so that successive pairs of pulses arriving at the cross correlator had a slightly different relative delay—in this way a sequence of blue pulses mapped out the cross correlation, with the resolution set by the frequency offset.

The 400 mW Ti:sapphire laser generated 14 fs transform-limited pulses at 78 MHz. By adjusting its cavity length, the difference in repetition rates between the cavities was set to $\Delta f = 2.8$ kHz, which produced a time resolution of 466 fs. The output of both oscillators was difference-frequency mixed in a 4-mm-long BBO crystal where the acceptance bandwidth of the BBO further limited the time resolution of the system to $\sim$1 ps. The cross-correlation signal was then filtered by a bandpass filter and measured with a photomultiplier (Hamamatsu R928). The peaks of successive mixed pulses is shown in the inset of Fig. 3, and the width of the cerium pulse can be retrieved from this trace.

The pulse duration of the cerium laser is shown in Fig. 3 as a function of the cerium cavity length detuning. We observed that the region where the oscillator generated the shortest pulses corresponded to the maximum cavity elongations of 20 μm from perfect synchronization. Moving away from this region, the pulses were not detectable with the difference-frequency-generation cross-correlator arrangement owing to the reduced peak power of the pulses.

The output spectrum shown in Fig. 4 was measured with an Ocean Optics HR4000 spectrometer and corresponds to the shortest pulse duration achieved (6 ps). The peak wavelength was at 291.7 nm, with a bandwidth greater than 1 nm. The wavelength of the sharp edge of the emission spectrum varied with alignment and pump power and so appears to be a genuine response to the decreasing gain

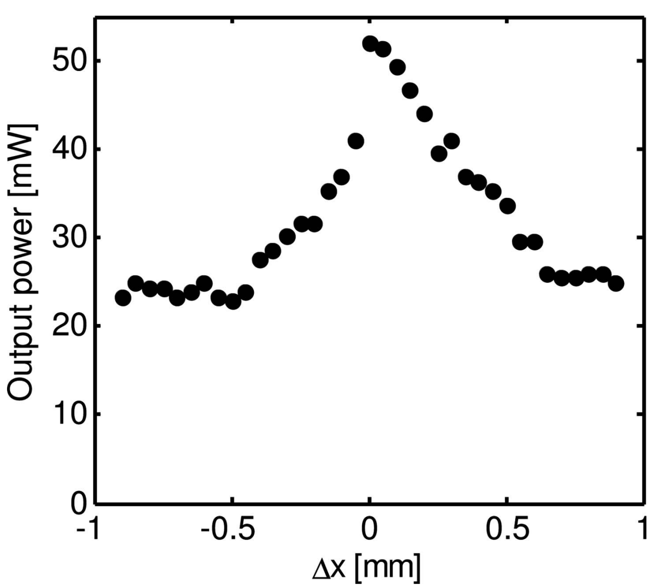

Fig. 2. Output power of the cerium oscillator as a function of cavity length detuning.

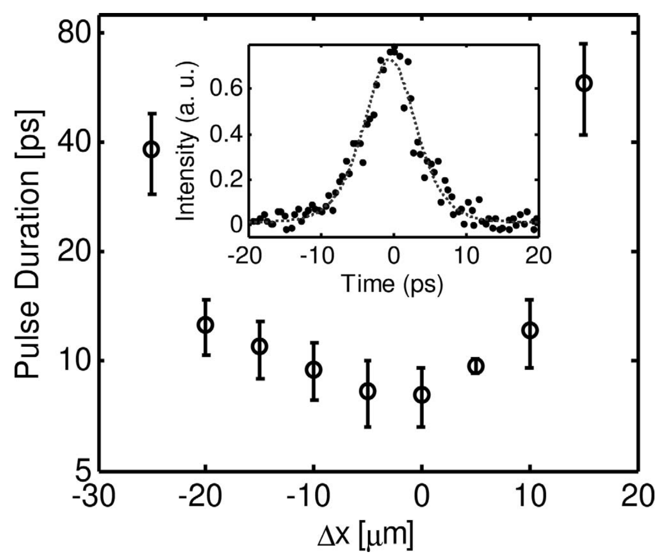

Fig. 3. Output pulse duration as a function of cavity length detuning. Inset, cross-correlation data for the measurement of the output pulse for zero detuning.

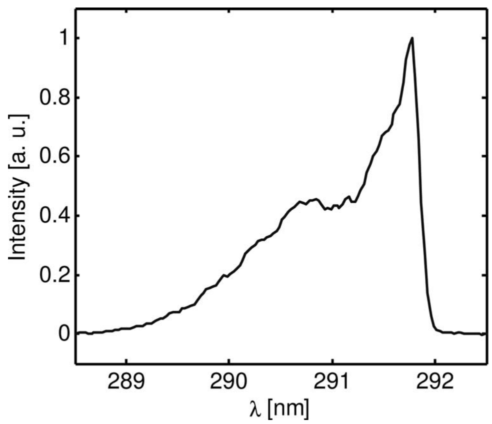

Fig. 4. Output pulse spectrum for zero detuning.

for longer wavelengths rather than an artifact from an unwanted absorption edge. Note that this bandwidth can support pulses of less than 100 fs, and so our pulses are not close to transform limited.

To achieve efficient operation of this low-gain system, reducing losses was vital; we used a three-mirror cavity rather than the more standard four mirror *z*-fold cavity to reduce the number of mirror reflections. Each reflection had 0.5% absorption loss, which is typical for high-quality dielectric mirror coatings in the deep UV. The Ce:LiCAF crystal also introduced approximately 0.5% scattering losses per pass, raising the total round trip loss to 3%. The measured slope efficiencies of 13%, with the optimized 3% output coupling, is consistent with the estimated losses, since nanosecond Ce:LiCAF lasers may operate with up to 46% slope efficiency [17]. We calculated the theoretical cw lasing threshold using Eq. (13) in [13]. Assuming a 14 μm waist diameter for the cavity mode, the value for the predicted absorbed pump threshold is 600 mW, in excellent agreement with our experiments (580 mW).

In conclusion, we have demonstrated what we believe to be the first mode-locked ultrafast cerium oscillator, generating output in the deep UV. The minimum pulse duration achieved was 6 ps, with a maximum output power of 52 mW. Our results represent the first major step to achieving few-femtosecond UV pulses directly from an all-solid state laser oscillator. We anticipate that with appropriate dispersion compensation and by using Kerr-lens mode locking the full ultrafast potential of Ce:LiCAF may be realized.

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