Cerium-Doped Fluoride Lasers

David W. Coutts and Andrew J. S. McGonigle

***Abstract*—In the 30 years since tunable ultraviolet (UV) lasers based on 5d → 4f transition of trivalent lanthanides doped into solid-state hosts were first demonstrated, tremendous progress has been made in these unique laser systems. Today, cerium-doped fluoride lasers offer wide tunability (280–333 nm), high efficiency (up to 62%) and narrow-band output. These lasers can also be used for femtosecond pulse amplification in the UV. Cerium lasers represent a logical route to generation of tunable UV in all-solid-state systems. In this paper, we review the current state-of-the-art cerium laser crystal development and cerium laser systems.**

***Index Terms*—Laser tuning, lasers, pulsed lasers, solid lasers, ultraviolet (UV) generation, ultraviolet (UV) spectroscopy.**

I. Introduction

TUNABLE ultraviolet (UV) laser sources are used in many applications including spectroscopy, remote sensing, and fluorescence detection. Traditionally, tunable UV output has been obtained from frequency-doubled dye lasers, frequency-tripled Ti:Sapphire lasers, or optical parametric oscillators (OPOs). In recent years, a new class of tunable UV laser has been developed based on cerium-doped fluoride crystals, which are tunable and lase directly in the UV when pumped with a suitable fixed-wavelength UV source. In this paper, the development and current state-of-the-art cerium lasers are reviewed in detail.

Elias *et al.* [1] first discussed the possibility of developing tunable UV or VUV lasers, based on the 5d → 4f transition of trivalent lanthanides doped into solid-state hosts, following observations of strong fluorescence arising from such materials [1]–[4]. Indeed, many such doped crystals were developed in the search for novel scintillator materials [5]. Unfortunately, in many of the potentially laser active materials excited-state absorption (ESA) from the upper laser level is stronger than the stimulated emission, leading to net loss. Thus, while there are two often cited reports in the literature [6], [7], in which VUV Nd^{3+} LaF_3 lasers are described, subsequent investigations and an examination of the presented data cast doubt on the evidence for VUV laser action in both cases. Indeed, in a comprehensive set of experiments using high quality Nd^{3+}:LaF_3 and Nd^{3+}:$LiYF_4$ crystals, including the same crystal used in [7], Cashmore *et al.* have demonstrated that ESA and color center formation leads to net loss in these materials when pumped with a VUV F_2 laser [8], [9].

Owen *et al.* [10] established necessary criteria for selecting suitable trivalent lanthanides ions and hosts, which have the potential for gain on the 5d → 4f transitions. The first criterion is that twice the expected laser frequency should not correspond to a photon energy for which there is normal absorption, in order to avoid ESA. The second criterion states that the lanthanide should have only one optically active electron in order to minimize the number of excited states, which could act as terminal states for ESA. Triple-ionized cerium, with its single 4f valence electron, is the only such trivalent lanthanide. The third criterion is that the host crystal should have the largest possible bandgap to reduce the possibility of ESA to the conduction band. This criterion suggests that fluorides (and possibly some borates and oxides), with their deep UV transparency, may be suitable hosts. Thus, the only tunable UV solid-state lasers which have been reported to date, are based on trivalent cerium-doped fluorides, including crystals such as Ce^{3+}:$LiCaAlF_6$ and Ce^{3+}:$LiLuF_4$, both of which have shown very efficient operation.

The absorption and emission characteristics of Ce^{3+}-doped fluorides are in many respects similar to laser dyes, with nanosecond upper laser level lifetimes and high quantum yields, and where color centers play an analogous role to dye triplet states. In common with dye lasers, cerium lasers have the advantages of high efficiency and excellent tunability; however, cerium lasers also have all the advantages of solid-state crystalline lasers including indefinite material lifetimes, simpler construction and compactness. Uniquely among solid-state lasers, cerium lasers produce output in the UV rather than the visible or IR. Thus we can obtain tunable UV in the 280–330 nm range directly from cerium lasers, without requiring frequency doubling/tripling of the tunable IR/visible wavelengths produced by all other tunable solid-state laser sources. Harmonic conversion of a visible or infrared pump laser may be required to match the cerium UV absorption bands; however, such a conversion is at a fixed wavelength and the relatively higher peak power of the pump laser leads to greater nonlinear conversion efficiency than harmonic up-conversion of a tunable visible/IR source. Furthermore, thermal effects are minimized by making the dissipative tunable laser stage the last frequency conversion process in the generation of tunable UV.

In light of the advantages listed above, considerable effort has been invested in searching for efficient, widely tunable laser active cerium-doped materials, and in developing laser technologies to exploit the possibilities of what has been described by one author [11] as a Ti:sapphire in the UV. In this paper, we review the development of cerium crystals and cerium laser technologies. The review is split into two sections. In the first sec-

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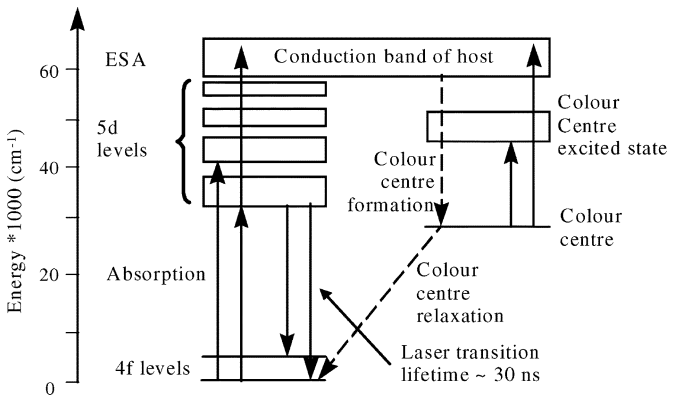

Fig. 1. Typical energy level structure for a trivalent cerium ion in a fluoride host.

tion, an overview of spectroscopic investigations aimed at characterising the gain, ESA, and color center properties of various cerium-doped materials (mostly fluorides) will be presented. In the second section, cerium laser device technology will be reviewed.

II. Spectroscopy

The free-ion ground configuration of Ce^{3+} consists of a xenon-like core of 54 electrons and a valence shell containing one 4f electron. This 4f configuration is split by the spin orbit interaction into $^2F_{7/2}$ and $^2F_{5/2}$ levels, separated by 2 253 cm^{-1} [12]. The first excited configuration of the Ce^{3+} free ion, formed by promotion of the 4f electron to the 5d energy level, is also split by the spin orbit interaction into the $^2D_{3/2}$ and the $^2D_{5/2}$ energy levels which lie at 49 737 and 52 226 cm^{-1}, respectively. The 6s level is located at 86 600 cm^{-1}.

Fig. 1 shows the energy level structure of Ce^{3+} when doped into a fluoride host. In the lattice, the outer 5s and 5p closed shells shield the 4f level from the crystal field. In this case, the weak field treatment of the crystal field interaction leads to Stark splitting of the $^2F_{7/2,5/2}$ manifolds, with typical Stark level splitting of $\sim$100 cm^{-1}. Conversely, due to the large spatial extent of the 5d wavefunction, beyond the 5s and 5p orbitals, the crystal field interaction dominates over the spin orbit interaction, depressing and splitting the 5d configuration into a number (typically 4 or 5) of broad Stark levels, each separated by $\sim$5 000 cm^{-1}. The number and position of the Stark levels are dependent on the strength and symmetry of the crystal field (for a detailed discussion of 5d levels in fluoride crystals see [13] and [14]).

UV lasing occurs on the 5d $\rightarrow$ 4f transition, in contrast to the IR 4f $\rightarrow$ 4f transition commonly employed in other trivalent lanthanide laser schemes such as Nd:YAG. While transitions within the 4f manifold are only permitted by mixing of opposite parity orbitals into 4f states by odd crystal field components, the 5d $\rightarrow$ 4f transitions in rare earth ions are electric dipole allowed. Thus, the 5d $\rightarrow$ 4f transitions have far higher electric dipole matrix elements and correspondingly smaller radiative lifetimes (a few tens of nanoseconds versus hundreds of microseconds) than the familiar IR transitions. Due to the large energy gap between the Ce^{3+} laser levels (typically 20 000 to 30 000 cm^{-1}), the probability of multi-phonon related nonradiative decay is low [15], resulting in high ($>\sim$90%) quantum efficiencies [14], [16].

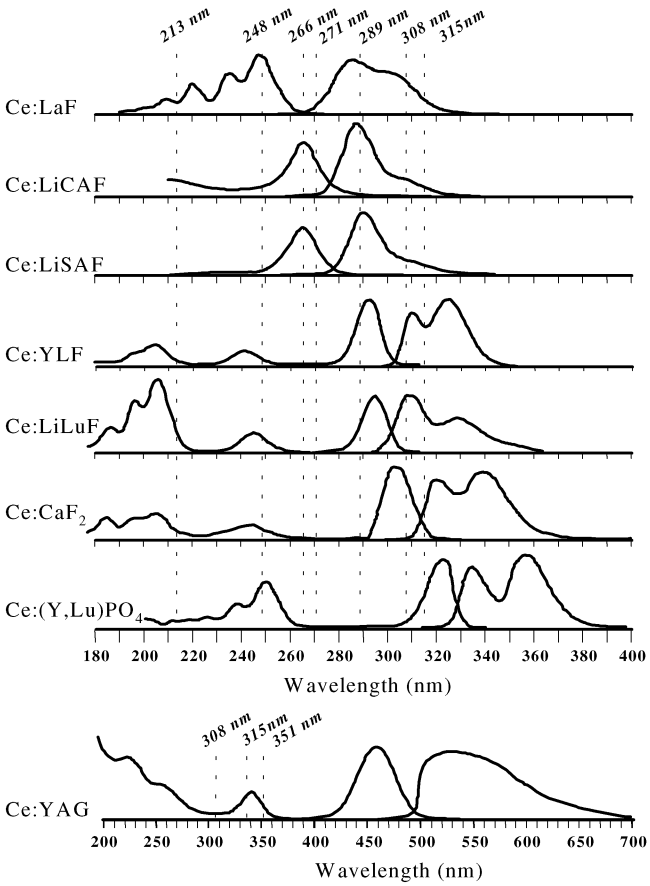

Fig. 2. Absorption and emission spectra for trivalent cerium in LaF [23], LiCAF [21], LiSAF [21], YLF [18], LiLuF [26], CaF_2 [24], $(Y,Lu)PO_4$ [27], and YAG [25], [28].

In some hosts, transitions involving higher lying excited states of the cerium ion, the conduction band and color centers may all play important roles in the UV spectroscopy [17]. The 5d $\rightarrow$ conduction band ESA transition and a color center formation mechanism, which is important for some of the Ce^{3+}-doped fluorides are shown in Fig. 1. Color centers may form when an electron, promoted to the conduction band following an ESA event, is trapped at a lattice defect or impurity site. Color center lifetimes vary with impurity trap depths, and with temperature, as they are thermally deactivateable. Color centers can potentially absorb at pump or laser wavelengths causing losses. Such absorption may promote the trapped electron to bound states of the trap or promote the electron back to the conduction band, whereupon the electron may return to a Ce^{3+} site, removing the color center.

Absorption and emission spectral regions, along with pump laser wavelengths for a selection of Ce^{3+}-doped materials are shown in Fig. 2. Broad absorption bands result from transitions from the 4f ground state to the crystal-field split 5d levels. Two peaks exist in the emission spectrum because transitions from the broadened, lowest 5d level terminate in both the 4f($^2F_{5/2}$) and 4f($^2F_{7/2}$) Stark manifolds. Emission is not observed from higher lying 5d levels because of fast multi-phonon relaxation to

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TABLE I
SPECTROSCOPIC CHARACTERISTICS OF SOME CERIUM-DOPED MATERIALS

Material	5d level lifetime (ns)	Emission cross section ($\times 10^{-18}$ cm^2)	ESA cross section ($\times 10^{-18}$ cm^2)	Remarks
Ce:YLF	40 [18]	7.6 unpolarized at 325 nm emission peak [18]	unknown	Stable and transient (50 ns, 200 μs and 50 ms) color centers, poor quantum yield.
Ce:LiLuF	40 [19]	6.7 π, 4.5 σ at 308 nm, [20]	<0.1 π, 2 σ at 308 nm [20]	ESA greatly reduces laser efficiency for σ polarization, but not π polarization.
Ce:LiCAF[a]	25 [21]	9.6 π, 6.2 σ at 290 nm emission peak [21]	3.6 π, 2.2 σ at 290 nm emission peak [21]	Gain cross section 6 $\times 10^{-18}$ cm^2 at 290 nm.
Ce:LiSAF[a]	28 [21]	9.5 π, 6.1 σ at 290 nm emission peak [21]	2.7 π, 4.6 σ at 290 nm emission peak [21]	Formation of color centers limits performance; however, use of an additional antisolarant pump can bleach color centers and restore efficiency [22].
Ce:LaF_3	20 [1]	unknown	unknown	Color centers limit performance at above 0.03 Hz [23]
Ce:CaF_2	40 [24]	unknown	unknown	Formation of color centers prevents lasing
Ce:YAG	65 [25]	unknown	up to 7 at 700 nm [25]	ESA prevents lasing

Typical dopant concentrations range from 0.01 to 2%at Ce^{3+}.
[a]Ce:LiCAF and Ce:LiSAF often grown with up to 2% Na^+ co-doping for charge compensation.n.

the lowest 5d level. The spectroscopic characteristics of several key Ce^{3+} materials are presented in Table I and discussed in the following sections, beginning with Ce:YAG.

A. Ce:YAG

One of the first materials to be investigated as a potential cerium laser was Ce^{3+}:$Y_3Al_5O_{12}$ (Ce:YAG), which was of particular interest because of its potential visible tunability. Unfortunately, laser activity was not demonstrated in this material. However, in the process of these investigations key insights into the selection of suitable hosts for cerium lasers were revealed.

In Ce:YAG the Ce^{3+} ion substitutes for the Y^{3+} ion at sites of D_2 symmetry, splitting the 5d level into five Stark manifolds [10]. The separations between the Ce:YAG laser levels are relatively small ($\sim$22 000 cm^{-1} cf. $\sim$35 000 cm^{-1} for Ce^{3+}-doped fluorides) because the 5d level is more split in oxide hosts than in fluorides. Thus, in contrast to the UV emission spectra observed from Ce^{3+} fluorides, the Ce:YAG emission spectrum peaks in the visible ($\sim$550 nm), as shown in Fig. 2.

Strong ESA to the conduction band prevents lasing in Ce:YAG [29]. This absorption has been observed in several studies, including a variety of pump–probe experiments, where pump pulses always induced net absorption with a lifetime of 65 ns, corresponding to the 5d level fluorescence lifetime. The strength of this ESA absorption was independent of excitation wavelength for pump lasers matching the lowest three absorption bands (pump wavelengths of 500 [25], 351/353 [28], 337 [10], [30], and 308 nm [25]). The ESA spectrum extends from 480 to 880 nm with a relatively high peak ESA cross section of 1×10^{-17} cm^{-2} [25]. The high ESA cross section indicates that ESA is an allowed transition that does not terminate in a higher lying 5d level (forbidden due to parity considerations). ESA must therefore terminate in the conduction band. Indeed, the conduction band edge derived from this ESA spectrum (30 440 cm^{-1} above the 4f level) coincides well with the threshold for one photon photo-ionization (30 650 cm^{-1}) [31]. ESA losses in Ce:YAG were also found to be temperature dependent. For example, Jacobs *et al.* [28] observed that by cooling a Ce:YAG crystal to below $-20\,^{\circ}$C ESA losses balanced the stimulated emission induced gain, leading to no net loss.

In addition to ESA, the formation of color centers is important in Ce:YAG. The second 5d level also corresponds to the onset of the conduction band, hence, UV pump lasers (pump wavelengths $<\sim$360 nm) can pump directly into the conduction band generating free electrons, which may form color centers [25]. For blue-green laser pumping into the lowest 5d band, subsequent ESA at visible and IR wavelengths can also lead to the formation of color centers (via the conduction band). The color centers have relatively long lifetimes ($\sim$10 μs) and relax via the 5d manifold [10]. Transient and permanent color centers may be thermally deactivated as observed by heating a LN_2 (77 K) cooled Ce:YAG crystal [10].

While gain has not been achieved in any of the Ce:YAG investigations, they serve to highlight the importance of choosing hosts that provide large 4f to conduction band energies to mitigate against ESA, and the formation of color centers. Indeed, all but one of the Ce^{3+} laser materials reported to date have been doped fluorides, which have characteristically large band gaps. Not all fluorides are necessarily good hosts, however. For example, Ce:CAF is also plagued by such severe color center formation, that gain will probably never be achieved in this material [24], [29], [32].

B. Ce:YLF and Ce:LiLuF

$Ce^{3+}LiLuF_4$ (Ce:LiLuF or Ce:LLF) and its isomorph $Ce^{3+}YLiF_4$ (Ce:YLF) were the first cerium materials in which lasing was demonstrated with reasonable (>1%) efficiency [8], [19]. Subsequently, Ce:LiLuF has demonstrated widely tunable (>30-nm tuning range [33]) and efficient (up to 62% slope efficiency [34], [35]) laser performance. These two materials are spectroscopically very similar, and are hence considered together in this section.

The Ce^{3+} ion substitutes for Lu^{3+}/Y^{3+} in Ce:LiLuF/Ce:YLF, resulting in fivefold splitting of the 5d level, with the lowest three absorption peaks situated at $\sim$207, 245, and 295 nm, and emission spectrum peaks at $\sim$310 and 327 nm [18], [19]. Both Ce:YLF and Ce:LiLuF have a 40-ns 5d level lifetime [18], [19], however, Ce:LiLuF has roughly twice the quantum yield of Ce:YLF (0.88 ± 0.08 [19]). Measurements of gain in a four pass Ce:LiLuF amplifier by Sarakura *et al.* [36] indicate that the saturation fluence of

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this material is of order 50 mJ/cm^2 (cf. $\sim$1 mJ/cm^2 for a dye laser), corresponding to an unpolarized emission cross section $\sigma_{\rm em}$ of $\sim 10^{-17}$ cm^2 at 325 nm.

Both Ce:LiLuF and Ce:YLF are affected by color center formation, however, this problem is considerably worse in the latter case [8], [19], [37]. In pump probe studies, absorption due to both transient and long lived color centers was much greater in Ce:YLF than for Ce:LiLuF [27]. Ce:YLF is found to form three classes of color centers with lifetimes of 50 ns, 20 μs, and 50 ms at room temperature, corresponding to different trap depths [38], [39] (cf. 40 ns 5d lifetime). These color centers' lifetimes decrease with increasing temperature, indicating thermal deactivatability. Color center formation is also a function of crystal purity. With high-quality Czochralski grown crystals, Ce:LiLuF color center absorption has sufficiently reduced that lasing with slope efficiencies in excess of 50% has been achieved [33], [40], [41]. Recently, Yb^{3+} (1%) codoping of Ce:LiLuF has further reduced the effects of color center formation, leading to operation of a Ce:LiLuF laser with 62% slope efficiency [34].

Both gain and ESA (and subsequent color center formation) are strongly polarization dependent in Ce:LiLuF [8], [42], [43]. Johnson [20] determined that the ESA cross section $(\sigma_{\rm ESA})$ for σ polarization (of order 2×10^{-18} cm^2 across the full tuning range of Ce:LiLuF) is more than an order of magnitude greater than the π-polarized ESA cross section. For the short wavelength tuning peak ($\sim$307 nm) the gain is highest for π polarization ($\sigma_{\rm em}(\pi) = 6.7 \times 10^{-18}$ cm^2 versus $\sigma_{\rm em}(\sigma) = 4 \times 10^{-18}$ cm^2), hence Ce:LiLuF lasers operate best in π polarization when tuned around this spectral peak [20]. At the long-wavelength peak ($\sim$327 nm) the emission cross section is higher for σ than π polarization ($\sigma_{\rm em}(\pi) = 3.7 \times 10^{-18}$ cm^2 versus $\sigma_{\rm em}(\sigma) = 6.8 \times 10^{-18}$ cm^2), as is the effective cross section $(\sigma_{\rm eff} = \sigma_{\rm em} - \sigma_{\rm ESA})$ which determines the gain. While the gain at 327 nm is highest for σ polarization, the relatively large ESA cross section for σ polarization leads to inefficient laser operation for this polarization. Therefore, the efficient operation of Ce:LiLuF lasers tuned around the long wavelength peak requires both a π-polarized pump and a polarization-biased cavity to ensure π-polarized output [20]. The harmful effects of ESA (and formation of color centers) in Ce:LiLuF are also found to be reduced by cooling the crystal from room temperature to $-3\,^\circ$C, even for π-polarized operation where a 60% improvement in output power was observed for 307 nm, π lasing [33].

C. Ce:LiSAF and Ce:LiCAF

To date, the two other cerium laser materials shown to demonstrate efficient lasing are Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) and its isomorph Ce^{3+}:$LiSrAlF_6$ (Ce:LiSAF). Ce:LiCAF appears to be the better of the two, with $>$35 nm tunability and $>$45% slope efficiency reported [44]–[46]. As with Ce:LiLuF and Ce:YLF, Ce:LiCAF, and Ce:LiSAF are very similar spectroscopically, thus they will be considered together in this section.

The 5d level lifetimes of Ce:LiCAF and Ce:LiSAF materials are 25 and 28 ns, respectively [21]. The quantum yield of Ce:LiCAF is $>$ 90%[16]. Both of these materials have absorption bands well matched to the 266-nm fourth-harmonic Nd:YAG wavelength (Fig. 2), and have a dominant emission peak around 290 nm, with a secondary peak around 309 nm.

Marshall *et al.* [21] measured small signal gain in Ce:LiCAF and Ce:LiSAF to determine absorption, emission and ESA cross sections (see Table I). They showed that the ESA cross section at the gain peak (290 nm) is higher for σ polarization ($\sigma_{\rm ESA} = 4.6 \times 10^{-18}$ cm^2) than for π polarization ($\sigma_{\rm ESA} = 2.7 \times 10^{-18}$ cm^{-2}) for Ce:LiSAF. They proposed that this disparity arises from the layered crystals' structures, in which the 5d $\rightarrow$ conduction band energy perpendicular to the c-axis (determined by Sr^{2+}/Ca^{2+} electronic wavefunctions) is lower than that for the π polarization (due to Li^+, Al^{3+} electronic wavefunctions). This theory also accounted for the σ-polarized ESA cross sections (at 290 and 266 nm) being greater for Ce:LiSAF than for Ce:LiCAF, because the spatial extent of the Sr^{2+} electronic wavefunction is greater than that for Ca^{2+}. Anisotropy is also evident in the polarized emission spectra, and the gain spectra [21] (e.g., Ce:LiSAF π-polarized $\sigma_{\rm EFF}$ is 6.8×10^{-18} cm^2 versus 1.5×10^{-18} cm^2 for σ polarization).

Ce:LiCAF appears to be a superior material to Ce:LiSAF in part due to its lower propensity to form color centers. Both transient (75 ns and 0.3 s lifetime) and permanent color centers have been observed in Ce:LiSAF lasers when pumped at 266 nm, while identical pumping conditions did not produce color centers in Ce:LiCAF [47]. As with many cerium laser materials, color center formation in Ce:LiSAF depends on crystal quality, particularly concerning the presence of impurities [21]. In Ce:LiCAF and Ce:LiSAF the Ce^{3+} ions substitute for the Ca^{2+}/Sr^{2+} octahedral sites leading to a charge imbalance. Bayramian *et al.* [22] found that codoping a Ce:LiSAF crystal with charge compensating Na^+ ions appreciably reduced color center formation, leading to an increase in laser slope efficiency from 17% (2% Ce:LiSAF) to 33% (2% Na, 2% Ce:LiSAF).

While Ce:LiSAF is easier to grow than Ce:LiCAF, large (11 cm) diameter Ce:LiCAF crystals have been grown recently by a Czochralski system in the Fukuda Laboratory Japan [11]. To improve crystal growth, mixed composition Ce:$LiSr_xCa_{1-x}AlF_6$ (LiSCAF) crystals have been developed as reported by Castillo *et al.* ($x = 0.0, 0.2, 0.35, 0.5, 0.65, 0.8$, and 1.0) [48] and Liu *et al.* ($x = 0.8$) [49]. Mixed composition Ce:LiSCAF crystals exhibit 25% longer 5d lifetimes than Ce:LiCAF, and the Ce:LiSCAF absorption is better matched to the 266-nm fourth-harmonic Nd:YAG wavelength than Ce:LiCAF [49].

D. New Ce^{3+}-Doped Laser Materials

Recently, several new Ce^{3+}-doped crystals have been reported, where gain is made possible by codoping with Yb^{3+} ions to reduce the deleterious effects of color center formation [27], [50]. Laroche *et al.* [27] report 25% single-pass gain at 355 nm for an absorbed 315-nm pump fluence of 0.18 J/cm^2 in Yb^{3+}-codoped Ce^{3+}:$LuPO_4$ (Ce:LuPO), the first report of gain in a cerium-doped oxide (phosphate). Semashko *et al.* [50] report lasing at 362 nm in a Yb^{3+} codoped Ce^{3+}:KY_3F_{10} (Ce:KYF) crystal pumped by a 308-nm XeCl excimer laser. These new results are highly significant, as these new materials have the potential to greatly extend the tuning range of cerium lasers beyond the present long wave tuning limit (335 nm for Ce:LiLuF) out to 370 nm and possibly into the blue (Fig. 2).

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TABLE II
KEY PERFORMANCE CHARACTERISTICS OF DIFFERENT CERIUM LASERS

Material	Pump λ's (nm)	Tunability (nm)	Output Power/ Energy	Maximum Efficiency
Ce:YLF	248	325 [18]	1.5 mJ [19]	5% (A) [19]
Ce:LiLuF	248, 289, 290	305-333 [33]	380 mW [43] 27 mJ [40]	62% (S) [34] 55% (S) [34]
Ce:LiCAF	263, 266, 271	280-316 [45]	900 mW 60 mJ [53] 98 mJ [11] 30 GW[a] [54]	46% (S) [46] 42% (A) [46]
Ce:LiSAF	263, 266, 271	283-313 [47]	250 mW [55] 5 mJ [47, 56]	47% (S)[b] [22]
Ce:LaF	248	286 [23]	5 μJ [23]	0.01% (A) [23]
Ce:KYF	308	362 [50]	-	-
Ce:LuPO	315	355[c] [27]	-	-

A = absolute efficiency, S = slope efficiency.
[a]Peak power from chirped-pulse amplifier system.
bPumped at 266 nm with additional 532 nm pump to bleach color centres
[c]25% gain measured at 355 nm.

Laser action was also reported for Ce:LaF_3 [23] in 1980; however, to date, Ce:LaF_3 lasers have only operated with low absolute efficiency (0.01% when pumped by a KrF laser). The poor efficiencies were attributed in part to poor crystal quality. It is not clear whether improved Ce:LaF_3 laser performance could be attained with higher quality crystals [51].

Upconversion pumping of Pr^{3+} codoped cerium materials has been investigated as a way to achieving UV lasing without requiring a deep UV pump laser, while avoiding color center formation [52]. As may be expected from the aforementioned conditions for achieving gain in 5d $\rightarrow$ 4f transitions [10], the addition of Pr^{3+} leads to ESA losses, which have outweighed any gain arising from energy transfer to Ce^{3+} in studies to date.

III. LASER TECHNOLOGIES

A summary of key cerium laser performance characteristics is presented in Table II.

A. Ce:LiLuF Lasers

Ehrlich *et al.* reported the first operation of a cerium laser: a KrF pumped untuned Ce:YLF laser, which produced 1 μJ of 325.5-nm output from 300 μJ of absorbed pulse energy [18]. Subsequently, untuned KrF pumped Ce:YLF output has been scaled to over 1 mJ, by using higher quality crystals and better optimized cavity configurations [8], [19]. The latter two reports document saturation in Ce:YLF pulse energy with increasing pump fluence and a decrease in Ce:YLF pulse energy for pump pulse repetition frequencies (PRFs) of greater than 0.5–1 Hz (for fixed pump-pulse energy), implying the involvement of transient color centers.

Far greater efficiencies have been obtained from the analogous material, Ce:LiLuF. Laser action in Ce:LiLuF was first demonstrated by Dubinskii *et al.* in 1992 [37], [57]. During an inter-comparative study with Ce:YLF, Ce:LiLuF demonstrated lower lasing thresholds, higher efficiencies and greater photochemical stability against the formation of transient and stable color centers. The highest Ce:LiLuF pulse energy yet reported [40] is 27 mJ at 309 nm with 17% slope efficiency, using a 1-Hz unpolarized KrF pump laser. This result was obtained using a transversely pumped high-quality Czochralski-grown laser crystal, as opposed to the Bridgman–Stockbarger crystals used in the earlier studies.

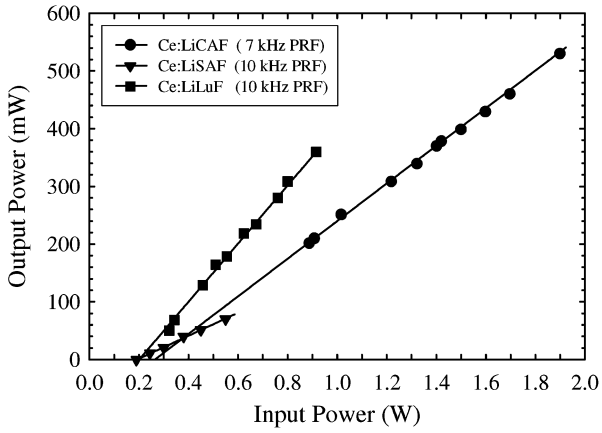

Fig. 3. Power scaling characteristics for Ce:LiCAF [45], Ce:LiSAF [59], and Ce:LiLuF [33] lasers all pumped by high-PRF frequency-summed/doubled copper vapor lasers.

To date, all KrF laser-pumped Ce:LiLuF lasers have been operated at PRFs of $\leq$ 20 Hz. However, many spectroscopic applications require tunable UV output at multi-kHz PRF. Operation at high PRF is more challenging due to the lower pulse energies available from high-PRF pump lasers and because of the increased possibility of color center population build up, with shorter inter-pulse relaxation periods. Multi-kilohertz PRF Ce:LiLuF laser operation has been investigated, using 289-nm frequency-doubled copper vapor laser (UV-CVL) pump sources. Such a laser (6.2-kHz PRF) was first reported by us [58], where we noted neither crystal coloration, nor decay in output following many hours of operation, in contrast to a previous study when the same crystal sample was pumped with a 248-nm KrF laser [8]. The improved performance may have been due to the π-polarized pump scheme; however, an additional advantage of the UV-CVL pump source is that the 289-nm wavelength pumps into the lowest 5d band, rather than the second-lowest 5d band accessed by 248-nm KrF lasers. By reducing the pump PRF to 60 Hz using a chopper wheel, the Ce:LiLuF pulse energy was increased more than twofold, indicating the tangible (if minimized) role color centers played in this laser scheme, which used a relatively poor quality crystal. Subsequently, 289-nm UV-CVL pumped untuned Ce:LiLuF powers have been scaled to 380 mW (at 309 nm), with slope efficiencies of 51% (see Fig. 3), [43]. These results, which are comparable to the best efficiencies obtained at lower (tens of hertz) PRFs, were obtained by cooling the crystal to $-3\,^{\circ}$C, which increased the crystal fluorescence intensity and 309-nm output power (by $\sim$50%), relative to room temperature operation. It was proposed that this enhancement arose from narrowing of the 5d $\rightarrow$ conduction band transition, thereby reducing ESA/color center formation. Recently, Semashko *et al.*

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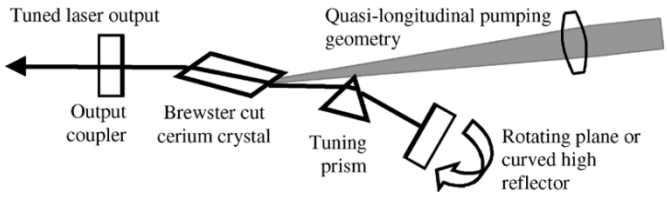

Fig. 4. End-pumped prism tuned cerium laser cavity configuration.

[34] have demonstrated highly efficient (62% slope efficiency) operation of a KrF pumped Ce:LiLuF laser incorporating 1% Yb^{3+}codoping of the active medium to reduce color center formation.

While KrF lasers and UV-CVLs are well matched to the absorption bands of Ce:LiLuF, for practical reasons, it is often desirable to develop all-solid-state laser technologies. However, unlike Ce:LiCAF and Ce:LiSAF, Ce:LiLuF cannot be pumped directly by the 266-nm fourth harmonic of an Nd:YAG laser. Several approaches to all-solid-state pumping have been investigated. Sarukura *et al.* [60] have used the fifth harmonic of an Nd:YAG laser (213 nm) to pump into Ce:LiLuFs 200–215-nm absorption band. This laser operated with very low efficiency ($\sim$2%), partly because of the poor quantum efficiency for 213 nm pumping, and possibly also due to exacerbated color center formation due to the short pump wavelength.

A more promising all-solid-state Ce:LiLuF laser was reported by Rambaldi *et al.* [41]. They used a 20-Hz 290-nm Ce:LiSAF pump source, itself pumped by the fourth harmonic of an Nd:YAG laser. In this laser scheme, as for the UV-CVL source, the Ce:LiSAF output pumps directly into the lowest 5d level, providing higher quantum efficiency and less potential for thermal problems or ESA/color center formation [41]. This system produced high (55%) slope efficiency in an untuned end-pumped configuration, with $>$ 2 mJ of 308-nm output from 6-mJ absorbed pump. Indeed, above 900-mJ/cm^2 pump fluence, the reflectivity of the crystal faces was sufficient to produce oscillation without any external cavity mirrors.

Recently, Johnson *et al.* [35] used a 289-nm 10-kHz PRF solid-state frequency-quadrupled Raman-shifted Nd:YAG laser to pump Ce:LiLuF. Up to 1.4 W of yellow output at 578 nm was generated from the Raman laser, from which up to 230 mW of UV at 289 nm was generated by frequency-doubling the yellow in beta-barium borate (BBO). The Ce:LiLuF laser gave 67-mW UV output with 62% slope efficiency (based on absorbed power). This slope efficiency matches the record 62% slope efficiency obtained by Semashki *et al.* using the new Yb^{3+} codoped Ce:LiLuF material, which is less susceptible to color center formation [34].

Broadly tunable operation of Ce:LiLuF lasers have been reported using cavities incorporating intracavity prisms (Fig. 4), and Littrow-prisms (Fig. 5). Low PRF tunability [40], [41], [60], results have been poor with tuning limited to the neighborhood of the two emission peaks (307.6–313 and 324–328.5 nm). This may be a result of poor crystal quality, but more likely reflects the use of unpolarized 248-nm KrF pump lasers, which exacerbate ESA and color center formation. We have reported continuous tunability for Ce:LiLuF lasers from 305 to 333 nm using a

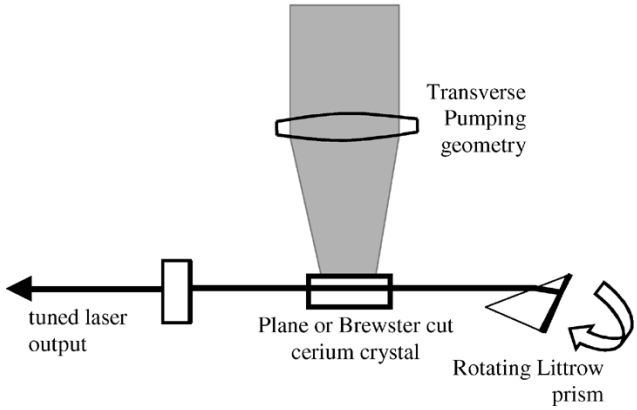

Fig. 5. Side-pumped Littrow prism tuned cerium laser cavity configuration.

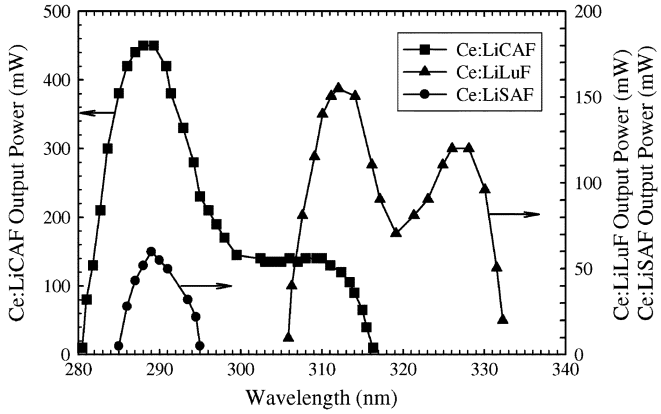

Fig. 6. Frequency tuning curves for Ce:LiCAF [45], Ce:LiSAF [59] and Ce:LiLuF [33] lasers all pumped by high-PRF frequency-summed/doubled copper vapor lasers.

10-kHz PRF UV-CVL pump laser (Fig. 6), [33]. In these experiments a quasilongitudinal pump geometry was used with a 2 mm long Ce:LiLuF crystal and a single silica Brewster prism (oriented to favor π-polarized oscillation) for wavelength tuning. Up to 150 mW of output was generated at the 311-nm tuning curve peak from 900 mW of pump power when the crystal was at room temperature. When cooled to $-2\,^{\circ}$C the output power increased by approximately 50% over the full tuning range, except around the 327-nm peak, where the output power decreased and changed from π polarization to σ polarization (even though the Brewster prism was oriented to favor π-polarized oscillation). At 327 nm, the gain is highest for σ polarization, but the efficiency is lowest due to the relatively large ESA cross section. Johnson *et al.* [20] found that inserting a polarizer to force π-polarized lasing restored continuous tunability.

Narrow-band operation of a 10-kHz UV-CVL-pumped Ce:LiLuF laser has been demonstrated by Johnson [61] where a prism expanded Littrow grating cavity was used to generate a few milliwatts of UV output with 0.7-cm^{-1} bandwidth and tunability from 306.5 to 311.5 nm.

B. Ce:LiCAF/LISAF Lasers

Dubinskii *et al.* [16], [62] are also credited with the first report of a Ce:LiCAF laser. A frequency-quadrupled Nd:YAG laser was used to longitudinally pump an untuned Ce:LiCAF

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laser. While the performance of this laser was modest (0.15 mJ of 288-nm output at 12.5-Hz PRF and ~9% slope efficiency), no color center induced discoloration of the Bridgman-Stockbarger grown crystals was evident following five hours of irradiation (1-J/cm^2 fluence), indicating good photochemical stability. In 1993, lasing was first reported in Ce:LiSAF, the isotype of Ce:LiCAF, benefiting from the relatively mature level of Cr:LiSAF growth technology [21], [63]. Subsequently, slope efficiencies of up to 39% [64], and pulse energies of 60 mJ (the highest yet achieved from any cerium laser) [53] have been realized, at PRFs of 10 Hz by using large (15 mm) diameter Czochralski-grown quasi-longitudinally pumped Ce:LiCAF crystals to allow scaled (4–6 mm) pump spot sizes. High (39%) slope efficiencies have also been obtained in a transversely pumped 10 Hz PRF Ce:LiCAF laser, where a Brewster cut crystal was used to favor the π polarization and hence minimize ESA and color center induced losses [65].

Ce:LiSAF appears to be slightly less efficient than Ce:LiCAF, with the highest reported slope efficiency of 33% [56], and pulse energies of up to 5 mJ reported from both longitudinally [56] and transversely [47] pumped 10-Hz PRF Ce:LiSAF lasers. However, by pumping a Na^+-codoped Ce:LiCAF crystal with an additional antisolarant 532-nm second-harmonic Nd:YAG beam, Bayramian *et al.* demonstrated that the color centers could be bleached, leading to increased laser slope efficiency (47%) [22].

High-PRF (multi-kilohertz) operation has been reported by several groups. Petersen *et al.* [55], [66] report Ce:LiCAF and Ce:LiSAF lasers longitudinally pumped at 266 nm from a 20-kHz PRF frequency-quadrupled Nd:YVO_4 laser. These LiCAF and LiSAF lasers gave average output powers of 350 and 250 mW and 28% and 27% slope efficiencies, respectively. Govorkov *et al.* [44] report a 1-kHz PRF Ce:LiCAF laser pumped by the fourth harmonic of an Nd:YAG laser. We have reported a 530-mW, 7-kHz PRF Ce:LiCAF laser [45] and a 70-mW, 10-kHz PRF Ce:LiSAF laser [59] both pumped by the 271-nm sum-frequency output from a CVL (see Fig. 3). In all these studies, the crystals Brewster cut to favor π-polarized oscillation were used. LiCAF lasers consistently outperformed LiSAF lasers, with $>$0.5 W average output powers obtained from Ce:LiCAF lasers in several of these studies ([44], [45]), and Ce:LiCAF laser slope efficiencies up to 32% [45]. Recently Fromzel and Prasad [46] have developed a 1-kHz PRF Ce:LiCAF laser with 46% slope efficiency and 600-mW output power pumped by a frequency-quadrupled Nd:YLF laser, and we have demonstrated 0.9-W average output power from a 10-kHz PRF Ce:LiCAF laser pumped with 4.1 W at 271 nm from a sum-frequency CVL laser.

Single-prism tunability of transversely and longitudinally pumped low PRF Ce:LiCAF and Ce:LiSAF lasers has been achieved in numerous investigations [44]–[47], [56], [59], [62], [63], [65]–[67], the broadest accomplished tuning ranges being 280–316 nm and 283–313 nm, respectively, in these media [44]–[47], [65]. Tuning curves for multi-kilohertz UV-CVL-pumped Ce:LiCAF and LiSAF lasers are shown in Fig. 6.

Typically, single prism linewidths are of order 0.3 nm for silica prisms and 0.1 nm for sapphire prisms [46]. Further linewidth narrowing can be achieved with use of an intracavity etalon and multiprism beam expander (see Fig. 7). Up to 130 mW of 290-nm 12.5-gHz linewidth output has been achieved with such a scheme [44]. Narrow linewidths have also been obtained from Ce:LiSAF using distributed-feedback (DFB) cavities, producing up to 80 μJ of 290-nm output, with a bandwidth of less than 0.1 nm [68]. Dubinskii *et al.* [69] have subsequently used such a distributed feedback Ce:LiCAF laser to injection seed a Ce:LiCAF ring laser. The seeded laser characteristics include: 0.015-nm output bandwidth, tunability from 282 to 302 nm, 8-mJ maximum output energy, and 32% slope efficiency.

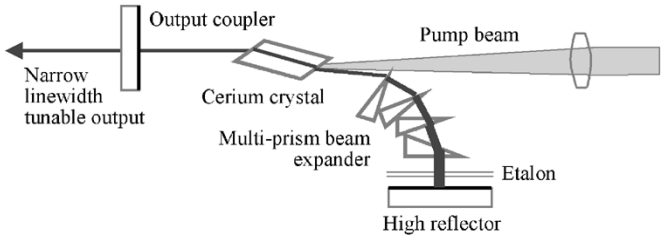

Fig. 7. Cavity configuration for narrow-band operation of a longitudinally pumped cerium LiCAF laser (after [44]).

Wavelength extension of the tunable Ce:LiCAF and LiSAF outputs has been achieved by sum-frequency-mixing with fixed wavelength lasers; typically, the infrared fundamental wavelength of the frequency-quadrupled Nd pump laser [55], [65], [67], [70]. For example, Pinto *et al.* [65] frequency summed the 1.046-μm Nd:YVO_4 line with output from a Ce:LiCAF laser (pumped by the same frequency-quadrupled Nd:YVO_4 laser), to produce tunable 228–240-nm output at 20-kHz PRF. Laroche *et al.* [71] have used Raman shifting in a 0.2-m-long high-pressure H_2 Raman cell to generate 4 Stokes and 2 anti-Stokes tunable outputs from a 10-Hz 1.4-mJ Ce:LiSAF laser (output wavelengths from 260 to 550 nm, although with large gaps in tunability). However, complete spectral coverage of this range based on second anti-Stokes to fourth Stokes conversion in hydrogen is theoretically possible for a combination of LiCAF and LiLuF lasers, provided they produce sufficient output energy over their complete tuning ranges to reach Raman conversion thresholds.

C. Short Pulse ($\leq$ 1 ns) Generation and Amplification

In addition to broad UV tunability, cerium lasers have considerable potential for ultrafast pulse generation and amplification [72]. In gain-switched operation, Sarukura *et al.* have obtained subnanosecond pulses from both Ce:LiCAF lasers [170-μJ, 700-ps full-width–half-maximum (FWHM)] [73] and Ce:LiLuF lasers (77-μJ, 880-ps FWHM) [60] by using short (2–3 cm) low Q-cavities and nanosecond pump lasers. By reducing the (266 nm) pump pulse duration to 75 ps, a further reduction of Ce:LiCAF laser pulse duration to 150 ps has been reported [67]. For the nanosecond-pumped systems, by adding an additional long (2.2-m round trip) feedback cavity to the short-pulse low-Q gain-switched laser cavity (Fig. 8), the same group has obtained trains of up to ten self-seeded subnanosecond pulses with an enhancement factor of up to 2% and $<$ 5% efficiency [74], [75]. Greater output powers and

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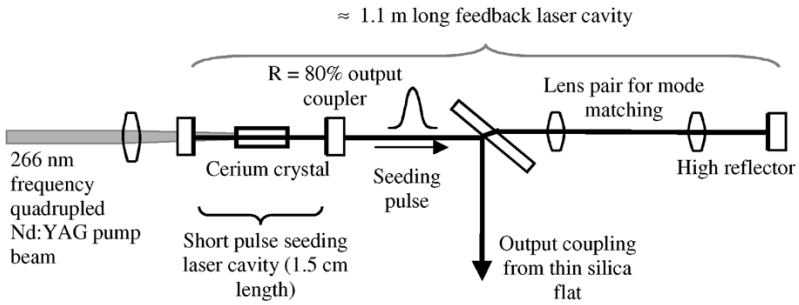

Fig. 8. Optical arrangement for short pulse cerium laser incorporating feedback cavity for pulse train generation (adapted from [74]).

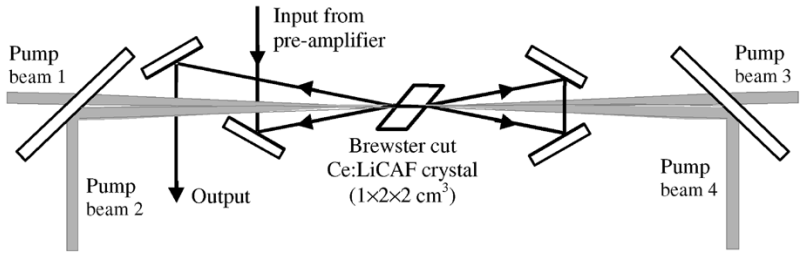

Fig. 9. Double-pass amplifier arrangement for high-energy pulse amplification in Ce:LiCAF with four 266-nm pump beams (adapted from [11]).

efficiencies may be achievable with high-Q feedback cavities combined with cavity dumping.

In an extension of the above short-pulse work, various multipass Ce:LiLuF [36] and Ce:LiCAF [11], [49], [54], [70], [76] amplifiers have also been developed by the Sarukura group [72]. Four pass Ce:LiLuF and Ce:LiCAF amplifiers, transversely pumped by a 10-Hz PRF KrF laser and longitudinally pumped by a 10-Hz PRF fourth-harmonic Nd:YAG laser, respectively, have been used to amplify short pulses. In the former case, amplifier gain of up to 17 dB, for a 3-ps 325-nm frequency-tripled mode-locked Ti:Al_2O_3 probe, was demonstrated [36]. In the latter case, the high-saturation fluence (115 mJ/cm^2) [21] of Ce:LiCAF was exploited to demonstrate chirped-pulse amplification of femtosecond pulses in a bow-tie style amplifier (with a gain of 370). Pulses of up to 6 mJ were derived, which were compressed down to 120 fs (2.5 mJ) using a grating pair compressor, or 115 fs (3.5 mJ) using a four-prism compressor [49], [54]. Using large Ce:LiCAF crystals (cut from 11-cm-diameter boules), a double-pass power amplifier (Fig. 9) has been developed by Ono *et al.* and used to generate up to 98-mJ 3-ns pulses at 290 nm [11] with 22% efficiency. Note that in addition to a record pulse energy, the average power from this 10-Hz system is also a record 0.98 W from any cerium laser or amplifier.

Elsewhere, Buhr and Fedosejeves [77] have designed and modeled a three-stage Ce:LiLuF chirped-pulse amplifier system with a predicted output of $>$ 1-J or 13-tW peak power, based on a 5.1-J e-beam-pumped KrF pump laser.

D. Summary of Laser Characteristics

A wide variety of efficient UV laser systems have been developed based on Ce:LiCAF, Ce:LiSAF, and Ce:LiLuF. Average output powers up to the 1-W level, output pulses of order 100 mJ, and $>$ 50% efficiencies all demonstrate that cerium lasers are practical devices with an important place, and, owing to their tunable UV outputs, certainly a unique place in the solid-state laser family.

To date, Ce:LiCAF and Ce:LiSAF have received the greatest attention of all the cerium laser media, partly because of pre-existing Cr:LiSAF growth procedures and also because these lasers, unlike Ce:LiLuF, can be readily pumped using 266-nm fourth-harmonic Nd:YAG lasers. Indeed, these are the only cerium laser crystals currently commercially available or used to date for laser applications (such as remote sensing of ozone and SO_2 [56] and combustion diagnostics [78], [79]). Based on the excellent tunability and efficiencies of Ce:LiCAF and Ce:LiSAF, it appears as though color center and ESA effects are of little significance when good quality crystals and π-polarized pump and laser modes are implemented. For these reasons, a Ce:LiCAF laser was brought to market by Lambda-Physik, as a replacement technology for frequency-doubled dye-laser UV applications [44].

Due to Ce:LiLuF's high quantum efficiency and good photochemical stability, this material operates with the highest slope efficiencies ($>$ 50%) [35], [41], [43] of any cerium laser. While color centers and ESAs are evident in this material, they can be minimized by implementing long ($\sim$290 nm) wavelength pump sources, π-polarized pump and laser modes and crystal cooling. Thus, in light of Ce:LiLuF's excellent performance characteristics, this material deserves wider recognition.

IV. Closing Remarks

This review is written approximately 30 years since the prospect of UV tunable cerium lasers, based on the 5d $\rightarrow$ 4f transition of trivalent lanthanides was first proposed and 20 years since the first two cerium lasers (Ce:YLF and Ce:LaF) were reported. A combination of ESA and color center formation prevented efficient operation during the early investigations (outputs $\leq$5 μJ), while completely quenching gain in subsequent studies of Ce:YAG and Ce:CaF. During the last 10 years, Ce:LiLuF, Ce:LiCAF, and Ce:LiSAF lasers, which are far less affected by ESA/color center effects, have been developed, recently demonstrating excellent performance characteristics (e.g., $>$ 50% slope efficiencies [41]; 280.5–316-nm tunability

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[45] and 60-mJ pulse energies [53]). Thus, cerium lasers are now available that rival, or exceed, the performances and efficiencies of second-harmonic dye and third-harmonic tunable solid-state lasers, providing direct UV tunability, without the complication of subsequent nonlinear frequency conversion. Despite these advances, cerium lasers have not received the recognition they merit. It is hoped that this article will prompt interest, both in further understanding the fundamental laser kinetics of cerium fluorides, and in the proliferation of these lasers for a host of UV spectroscopic applications, to which they are highly suited.

The quest to find additional laser active cerium fluorides is ongoing, and there have been a number of recent preliminary reports into the possibility of lasing in fluorescent media such as Ce:SAF_2 [80], Pr, Ce:YPO [81], Ce:LiBaF [82], [83], Ce:KYF and Ce:LiGdF [84], Ce:LiSGF [85], and Ce:KMgF [86]. However, as yet none of these materials have lased, thus the list of cerium lasers remains: Ce:YLF, Ce:LaF, Ce:LiLuF, Ce:LiCAF, Ce:LiSAF, with the very recent observation of gain in Ce:LuPO [27] and lasing in Ce:KYF [50]. Crystal growth technology continues to be refined in several laboratories [11], [48], [87], including growth of 11-cm-diameter boules [11].

To date, cerium lasers have been restricted to pulsed-mode operation. However, over the last few years, high-power (multiple Watt) continuous-wave (CW) fourth-harmonic Nd:YAG lasers have become available e.g., [88], opening possibilities for CW Ce:LiSAF or Ce:LiCAF lasers. This prospect has already been discussed by Marshall *et al.* [21], who noted that ESA would be relatively unimportant, due to the small population inversions involved in CW operation, estimating $\sim$12 kW/cm^2 lasing thresholds, for 99% reflective output couplers. CW operation of Ce:LiLuF lasers may also be possible, with the 244-nm line from a frequency-doubled argon-ion laser providing a suitable pump wavelength. Given that chromium-doped LiSAF can produce ultrashort IR pulses via mode-locking, it is also conceivable that femtosecond pulses could be generated directly in the UV from Ce:LiSAF [89] and the other cerium laser materials (note LiLuF and LiCAF have similar nonlinear optical properties [90]). The 35-ns gain bandwidth typical of cerium lasers is theoretically capable of supporting oscillation of sub-10-fs pulses.

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