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All-solid-state laser system tunable in deep ultraviolet based on sum-frequency generation in CLBO

S.B. Mirov [a,*], V.V. Fedorov [a], B. Boczar [b], R. Frost [b], B. Pryor [b]

[a] *Department of Physics, University of Alabama at Birmingham, Birmingham, AL 35294, USA*
[b] *Light Age, Inc Two Riverview Drive, Somerset, NJ 08873, USA*

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Abstract

An efficient, all-solid-state laser system continuously tunable over the entire 196–204 nm spectral region was demonstrated. This new deep-UV source is based on a novel alexandrite laser/alexandrite-laser-pumped LiF:F_2^{+**} color-center laser (CCL) system. Tunable UV generation was realized using type I sum-frequency generation in CLBO of a frequency tripled alexandrite laser and a tunable oscillation of LiF:F_2^{+**} CCL. The maximum conversion efficiency obtained at 198 nm wavelength was 23% for input pump power densities of UV ($\lambda_2 = 245$ nm) and IR ($\lambda_1 = 1036$ nm) radiation of 1.5 and 3.3 MW/cm^2, respectively, and 25 Hz repetition rate.

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High power laser sources with radiation wavelengths below 200 nm have attracted significant attention for use in photochemical processes, semiconductors and integrated circuit processing and medicine. Such deep UV is not available to a direct or harmonic output of a single solid-state laser due to phase-matching constraints of appropriate nonlinear crystals. The only exception is for sum-frequency mixing (SFM) in BBO crystal of the third and fundamental harmonic radiation ($\hbar\omega + \hbar 3\omega$) of alexandrite or Ti:sapphire lasers. However, this approach is impractical for high power regimes of operation due to a high BBO absorption band edge around 200 nm. Cesium lithium borate (CLBO) crystal is a newly developed nonlinear optical crystal from the borate group. It is unique in many aspects, especially in terms of high damage threshold, large phase-matching acceptance angle and wide transparent range. The crystal has a UV transmission cutoff wavelength at 180 nm, which is shorter than that of BBO. Sophisticated SFM multi-stage systems with IR to deep-UV up-conversion in CLBO crystal based on three Nd:YLF and Ti:sapphire MOPA systems has been recently realized [1]. It is much more promising than a BBO based system for high output power applications, however, being very sophisticated (six nonlinear crystals, three pump lasers, one tunable laser, precise timing control with

* Corresponding author. Tel.: +1-205-934-8088; fax: +1-205-934-8042.
E-mail address: mirov@uab.edu (S.B. Mirov).

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two pulse generators for temporal pulse overlap) it is hardly manageable in commercial settings.

In our paper we describe an efficient room temperature, all-solid-state laser system that is continuously tunable over the entire 196–204 nm spectral region. This new deep-UV source is based on a novel alexandrite laser/alexandrite-laser-pumped LiF:F_2^{+**} color-center laser (CCL) system. F_2^+ CC consists of an electron trapped by two adjacent anion vacancies. In recent studies of LiF:F_2^{+**} CCL [2–4], it was shown that the LiF: F_2^{+**} laser exhibits excellent photo and thermostable operation at room temperature when pumped by an alexandrite laser and can provide efficient high power lasing tunable in 800–1200 nm spectral range. The alexandrite-laser-pumped LiF: F_2^{+**} laser exploits the best features of both the high gain, "dye-like" CCL and the high energy storage alexandrite laser. Due to a high gain and short CCL built-up time the system exhibits virtually no temporal delay between pump and output pulses for the entire range of CCL tunability [3,4]. These results allowed us to build a reliable LiF:F_2^{+**}-alexandrite laser source for a number of efficient nonlinear processes, including harmonic, sum-frequency, and difference-frequency generation.

Tunable UV radiation was achieved by sum-frequency generation of the third harmonic of the alexandrite laser and the fundamental harmonic of LiF:F_2^{+**} CCL in a CLBO nonlinear crystal. A schematic diagram of the deep-UV SFG setup is depicted in Fig. 1. The alexandrite laser is a basic source of optical radiation and served simultaneously for CCL pumping as well as for frequency up-conversion to 240–260 nm UV spectral range. The alexandrite laser tunable in 720–800 nm spectral range operated in a Q-switched regime with a 25 Hz repetition rate, pulse duration of 60–100 ns, and pulse energy up to 80 mJ. For optimal four level laser operation the alexandrite rod was heated to 80 °C, however, in some experiments to improve the alexandrite-laser output at the short-wavelength operation (around 720 nm) the temperature of the rod was decreased to 50 °C.

The alexandrite-laser output radiation was partially reflected by a beamsplitter ($R = 55\%$) for LiF:F_2^{+**} CCL pumping. The rest of the radiation was used for third harmonic generation. A type I

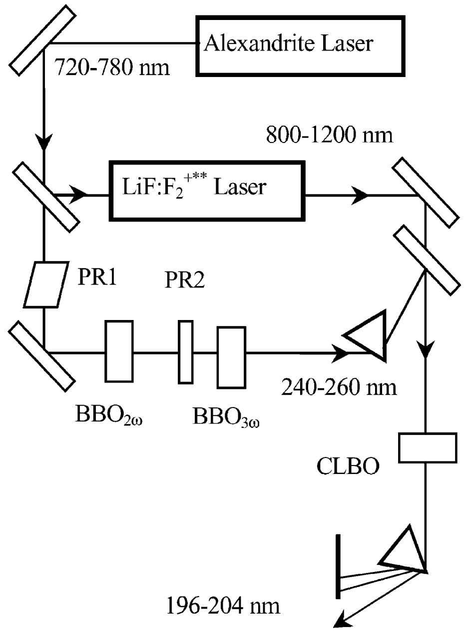

Fig. 1. Optical setup for SFG of deep-UV radiation.

(ooe) interaction in 10 mm length BBO crystals was utilized for generation of second ($\hbar\omega + \hbar\omega = \hbar 2\omega$) and third harmonics ($\hbar\omega + \hbar 2\omega = \hbar 3\omega$) of the alexandrite laser. The polarization rotator (PR2) positioned after frequency doubler served to compensate the difference between polarizations of the first and second harmonic radiations. The polarization rotator (PR1) ensured coincidence of polarizations of LiF:F_2^{+**} CCL and SHG. The tunable LiF:F_2^{+**} CCL (800–1200 nm) operated in a Littrow configuration [4]. It is based on an input dichroic mirror and diffraction grating (1200 grooves/mm) with diffraction efficiency of $R = 50\%$. A zero order of diffraction served for output. An unfocused pumping beam was introduced into 4 cm long LiF crystal through the input mirror. The LiF crystal doped with LiOH impurities was grown by the Kyropulos technique. To create LiF:F_2^{+**} active element, LiF crystals were γ-irradiated with a dose of 5×10^7 rad using a ^{60}Co source. The total tuning curve of the LiF:F_2^{+**} laser with optimum input mirrors under alexandrite-laser pumping is shown in Fig. 2 [4]. As one can see the tunable oscillation of LiF:F_2^{+**} laser extends from 800 to 1210 nm with maximum efficiency of

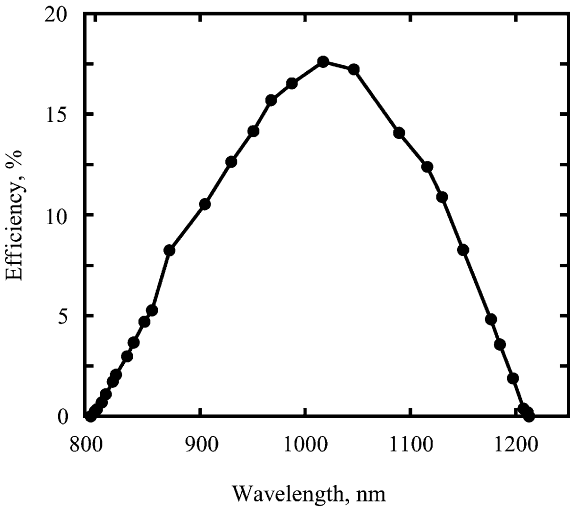

Fig. 2. Tuning curve LiF:F_2^{+**} laser under alexandrite-laser pumping ($\lambda_p = 740$ nm) at room temperature.

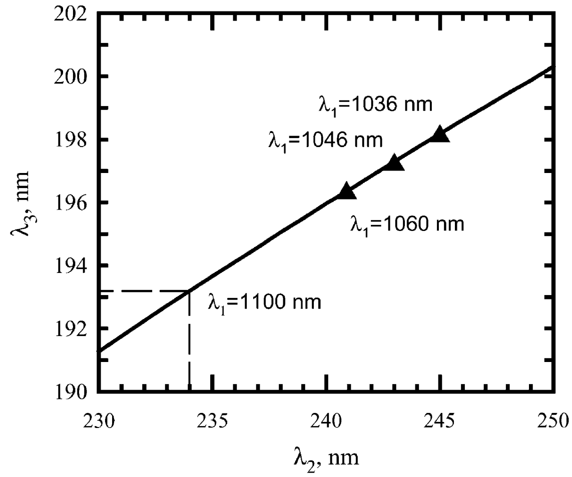

Fig. 3. Theoretically calculated (solid lines) and experimentally realized (filled triangles) CLBO 90° phase-matching dependences of SFG wavelengths (λ_3) versus pump wavelengths (λ_2).

17%. It is important to note that the temporal overlap of the pump and tunable radiations was smaller than the pump pulse duration and, hence, it does not require any additional time delay lines. The UV and NIR beams compressed to 1 mm in diameter were directed by the beam combiner to CLBO crystal where SFG occurred. The type I SFG process was realized in 10 mm long CLBO uncoated crystal cut for 90° noncritical phase matching ($\theta = 90°$, $\varphi = 45°$).

According to the early publications of Sellmeier's equation, CLBO crystal should have phase matching for SFG of 1064 nm and UV radiations around 237 nm with generation wavelength down to the crystal transmission edge at 180 nm [5,6]. However, new data on Sellmeier's equations [7] limits the shortest wavelength of SFG to 195 nm when 1064 nm pumping is used. In our study we performed experiments on measuring minimum wavelengths of LiF:F_2^{+**} CCL oscillation that correspond to 90° type I SFG noncritical phase matching in CLBO for different wavelengths of UV radiations of the third harmonic of alexandrite laser. Fig. 3 shows a variation of SFG wavelength for three UV wavelengths of 245 nm (THG of $\tau_\omega = 735$ nm), 243 nm (THG of $\lambda_\omega = 729$ nm), and 240.9 nm (THG of $\lambda_\omega = 722.8$ nm). The 90° phase matching was realized for CCL wavelengths of $\lambda_1 = 1036$, 1046, and 1060 nm, which corresponded to SFG, generated wavelengths of 198.1, 197.2, and 196.3 nm, respectively. The solid curve presents the theoretical results based on Sellmeier's equations refined in Ref. [7]. As one can see, there is a good agreement between the theoretical and experimental data. According to Sellmeier's equations [7], the shortest IR wavelength required for 193 nm generation is 1.1 μm. UV radiation that suits to this interaction should be less than 234 nm.

The deep-UV tuning curve of SFG is depicted in Fig. 4. The maximum SFG conversion efficiency at $\lambda_3 = 198.7$ nm was ~20–23% with respect to UV peak power of the third harmonic of the alexandrite laser. The short-wavelength part of the tuning curve represents SFM in a 90° noncritically phase matched CLBO crystal. The drop of the alexandrite-laser output energy at the wavelengths shorter than 730 nm results in low peak powers of the interacting UV and IR radiations and a corresponding decrease in the SFG efficiency at the short-wavelength tail of the tuning curve. The range of tunability can be further enhanced to 193 nm for 90° noncritical phase matching in CLBO with a reasonable efficiency, but it requires the output wavelength of alexandrite laser to be tuned to 701 nm ($233.66^{-1} + 1110^{-1} = 193^{-1}$). The long-wavelength part of the tuning curve is limited

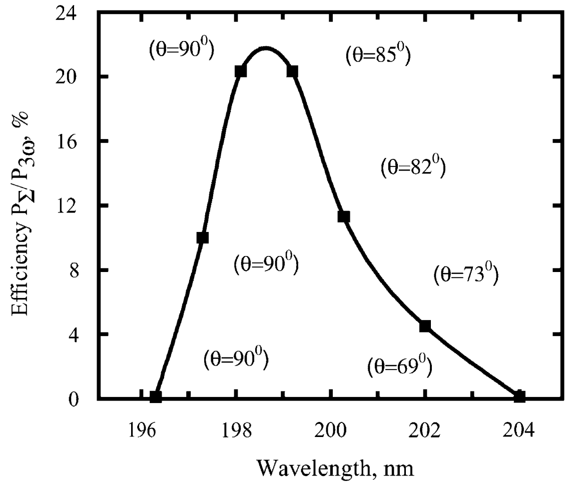

Fig. 4. Deep-UV tuning curve of alexandrite 3ω (241–247 nm)—LiF:F_2^{+**} (1030–1173 nm) sum-frequency generator based on CLBO crystal.

by the tuning range of the LiF:F_2^{+**} CCL. The conversion efficiency in our experiments was determined by available pump energy of alexandrite laser. The power densities of UV ($\lambda_2 = 245$ nm) and IR ($\lambda_1 = 1036$ nm) radiation were 1.5 and 3.3 MW/cm^2, respectively, which is considerably smaller than laser induced damage threshold for CLBO (26 GW/cm^2). Further 5-fold upscale of the alexandrite pump energy and peak powers of the interacting beams will result in a deep-UV tunability at an average power level approaching 1 W.

In conclusion we have demonstrated an all-solid-state laser system tunable in deep-UV range (196–204 nm) using alexandrite laser as single pump source. Tunable UV generation was realized using type I sum-frequency generation in CLBO of third harmonic of alexandrite laser and tunable oscillation of LiF:F_2^{+**} CCL. In our experiments, the spectral dependence of noncritical phase-matching correlates well with the new Sellmeier's relations [7]. The results show that CLBO can be used for 193 nm generation when pumping wavelengths are shorter than 237.6 nm. We have now produced a widely tunable UV light with maximum conversion efficiency of 23% obtained at 198 nm for input pump power densities of UV ($\lambda_2 = 245$ nm) and IR ($\lambda_1 = 1036$ nm) radiation of 1.5 and 3.3 MW/cm^2, respectively, and 25 Hz repetition rate. It appears likely that with improved operation of alexandrite laser similar simple sources producing watts average power can be developed.

References

[1] J. Sakuma, K. Deki, A. Finch, Y. Ohsako, T. Yokota, Appl. Opt. 39 (2000) 5505–5511.
[2] S.B. Mirov, T.T. Basiev, IEEE J. Special Top. Quant. Electron. 1 (1995) 22–30.
[3] A.Yu. Dergachev, S.B. Mirov, Opt. Commun. 145 (1998) 107–112.
[4] S.B. Mirov, A.O. Okorogu, W. Lee, D.I. Crouthamel, N.W. Jenkins, K. Graham, A.R. Gallian, A.Yu. Dergachev, W.B. Yan, W.J. Strachan, T.F. Steckroat, D.F. Heller, J.C. Walling, Proc. SPIE 3491 (1998) 1082–1088.
[5] N. Umemura, K. Kato, Ultraviolet generation tunable to 0.185 μm in $CsLiB_6O_{10}$, Appl. Opt. 36 (1997) 6794–6796.
[6] Y. Mori, I. Kuroda, S. Nakajima, T. Sasaki, S. Nakai, Appl. Phys. Lett. 67 (1995) 1818–1820.
[7] N. Umemura, K. Yoshida, T. Kamimura, Y. Mori, T. Sasaki, K. Kato, in: M. Fejer, H. Injeyan, U. Keller (Eds.), OSA Trends in Optics and Photonics, Advanced Solid State Lasers, vol. 26, Optical Society of America, Washington, DC, 1999, pp. 715–719.