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Taking advantage of light-emitting-diode (LED) performance breakthrough driven by the lighting market, we report, to the best of our knowledge, the first LED-pumped chromium-doped crystal laser oscillator and amplifier based on alexandrite crystals (Cr^{3+}:$BeAl_2O_4$). We developed a Ce:YAG concentrator as the pumped source, illuminated by blue LEDs that can be easily power scaled. With 2200 LEDs (450 nm), the Ce:YAG concentrator can deliver to the gain medium up to 268 mJ at 10 Hz at 550 nm with a irradiance of 8.5 k W/cm^2. We demonstrate, in oscillator configuration, an LED-pumped alexandrite laser delivering an energy of 2.9 mJ at 748 nm in free running operation. In the cavity, we measured a double-pass small signal gain of 1.28, which is in good agreement with numerical simulations. As an amplifier, the system demonstrated to boost a CW Ti:sapphire laser by a factor of 4 at 750 nm in eight passes with a large tuning range from 710 nm to 800 nm.

The first light-emitting-diode (LED) pumped laser is dated back from the mid-1960s [1]. At that time, LEDs had low efficiency and operated at cryogenic temperature (80 K). Therefore, this pump source has progressively been forgotten in the 1970s with the birth of laser diodes. Yet, since the mid-2000s LED performance at room temperature has experienced tremendous improvement; in parallel, their cost collapsed driven by the lighting market. Considering the lack of efficient and simple pump sources in the visible, LEDs can be today seriously reconsidered for laser pumping in this spectral range. Indeed, LEDs are more robust, less expensive, and much less sensitive to external environment than laser diodes for instance. Therefore, an LED-pumped laser source could benefit industrial, medical, and scientific laser applications.

In the last few years, direct LED-pumping for solid-state lasers focused on narrow bandwidth gain media. Typically Nd-doped materials that exhibit high gain [2–5] such as Nd:YVO_4 pumped at 850 nm (Barbet *et al.* [2]), Ce:Nd:YAG pumped at 450 nm (Villars *et al.* [3]), Nd:YAG pumped at 750 nm (Huang *et al.* [4]), and Nd:YAG pumped at 810 nm (Chen *et al.* [5]) have been demonstrated. If the different configurations exposed in the literature are leading to increasing optical efficiencies, LEDs' irradiance is still too low to pump transition-metal-doped gain media such as Ti:sapphire, Cr:LiSAF, or alexandrite. Indeed, these crystals are interesting for their broad emission spectrum and tunability, but their product "emission cross section *x* lifetime" is an order of magnitude lower than the Nd-doped-crystal one. Typically, LEDs would need a performance enhancement of an order of magnitude to reach the laser threshold in these gain media. This explains why LED-pumped transition-metal-lasers have never been reported ever since.

To meet this challenge, we propose to use luminescent concentrators (LC), which are able to improve the LED irradiance by an order of magnitude by a cost of wavelength conversion. Such concentrators are well known in the field of photovoltaics [6] and particle detection [7] and have now been recently used to pump Nd:YVO_4 laser and passively Q-switched Nd:YAG laser with 450 nm LEDs (2016 [8] and 2017 [9]).

As the first LED-pumped tunable medium, this work proposes to investigate alexandrite (Cr^{3+}:$BeAl_2O_4$). Indeed, pumped by flashlamps, this crystal has been used for decades in medical applications, especially in dermatology. In 1979, it was also the first tunable laser crystal directly diode-pumped at room temperature demonstrated by Walling *et al.* [10]. Its high absorption coefficients around 410 nm and 590 nm and long fluorescence lifetime (260 μs) enable flashlamp pumping [10] and red laser diode pumping. More recently, alexandrite laser sources were developed for remote sensing LiDAR systems [11]. And, pumped by high power red laser diodes, alexandrite laser reached an output energy of 23.4 mJ at 100 Hz in a slab configuration [12].

However, during the past 10 years, high-power red laser diodes have not followed the same development as near-infrared laser diodes: their power remains limited, and their cost is still high, typically in the range of few \$100 per watt. Considering that LEDs costs are below \$0.5 per watt and continuously decreasing, LED pumping of alexandrite arouses intense interest.

In addition, alexandrite tunability between 700 nm and 830 nm makes this crystal potentially attractive for femtosecond oscillators [13] and amplifiers. Alexandrite-based femtosecond amplifiers have been developed during the late '80s and '90s [14,15]. Indeed, spectral properties of alexandrite can be tuned versus temperature. A higher temperature induces a reduced fluorescence lifetime, a higher emission cross section, and, what is particularly interesting, a redshifted emission band [16] allowing an overlap with Ti:sapphire emission spectrum for instance. Considering the high cost of the green pump lasers used in Ti:sapphire femtosecond laser chains, LED pumped alexandrite amplifiers may have the potential to cut the cost of a laser chain by an order of magnitude.

In this Letter, an LED-pumping device *via* Ce:YAG luminescent concentrator is presented. It is used to pump an alexandrite laser oscillator in the mJ range. An intracavity double-pass small signal gain is measured and numerically corroborated. Power scaling is also demonstrated. Finally, the LED-pumped alexandrite is used in a tunable multipass amplifier for a continuous (CW) Ti:sapphire laser amplifier, the first step toward femtosecond pulse amplification.

First, the conception of the pump head relying on LEDs and Ce:YAG luminescent concentrator is described. As a primary pump source, blue LEDs with an emission spectrum centered at 450 nm (LUXEON *Z* Royal Blue from Lumileds) have been selected (see LED spectrum on Fig. 1). With a continuous drive current of 1 A, a unique LED emits an irradiance of 90 W/cm^2 for an emitting area of 1 mm × 1 mm. Pulsing the injected current with a square shape during 260 μs with a nominal current of 5 A, the peak LED irradiance increases to 315 W/cm^2. The LEDs' printed circuit board is mounted on a water-cooled heat sink.

In order to increase further LED irradiance, an Ce:YAG luminescent concentrator is used. Luminescent concentrator is a slab (14 mm × 1 mm × 100 mm in our case) proposing a large aspect ratio between the pump surface (14 mm× 100 mm) and the emitting surface (1 mm × 14 mm). The 450 nm light from the LEDs is absorbed by Ce^{3+} ions and reemitted within the concentrator (see Fig. 1 for Ce:YAG absorption and emission spectrum). The 550 nm light emitted in the concentrator is guided toward the edge of the concentrator *via* total internal reflections [8,9].

The concentration factor of the luminescent concentrator is the ratio of the luminescent concentrator output irradiance over the LED irradiance assuming that both sources have Lambertian emission. The concentration factor is driven by the LED filling factor on the concentrator pump surface and the aspect ratio of the luminescent concentrator. The thickness of the concentrator has been fixed to 1 mm, so that all blue light from LEDs is absorbed. The number of LEDs to pump our luminescent concentrator is 1120 (560 LEDs on each 14 mm × 100 mm surfaces). In this configuration, the filling factor is set to 41%. This value of the filling factor is the highest-ever reported in this kind of LC system [9]. This is the key parameter that permits the record concentration factor of our LC. Pulsing the LEDs at 10 Hz with a duration of 260 μs, the output power measured at the LC surface (1 mm × 14 mm) is 2.1 kW/cm^2 in air and 5.3 kW/cm^2 using an index-matching optical adhesive to couple the light in the laser crystal. The optical efficiency of the concentrator is then, respectively, 7.2% and 18% for a concentration factor of 6 and 15, respectively. This means that the LC increased the radiance of a LED by a factor of 15. The luminescent concentrator provides then 167 mJ (considering index matching between the LC and the gain medium) and can be used to pump an alexandrite crystal. In a broader perspective, this alternative light source is promising for pumping any crystal absorbing in the yellowish wavelength range where issues remain important due to the low efficiencies of laser diodes in this spectral range.

The pumping of alexandrite with this LED-pumped concentrator is now presented because the cerium emission band around 550 nm matches well the absorption band of alexandrite (Fig. 1). The alexandrite dimensions are 1 mm × 2.5 mm × 14 mm matching with the luminescent concentrator output facet (Fig. 2). They are bonded together using UV curing optical adhesive with a refractive index of 1.5. The laser crystal is 0.22 at.% Cr-doped and oriented such that the pump arrives on an alexandrite surface parallel to the *b* axis in order to have maximum absorption. One of the laser crystal longitudinal surfaces is coated to have high reflectivity over 660–750 nm, the opposite surface being AR coated for the same wavelength range. We design a plano–concave, 44 mm long laser cavity using 300 mm radius-of-curvature output couplers with different transmissions: 1.0%, 2.0%, and 3.3% at 750 nm.

The LEDs are pulsed for 260 μs to match alexandrite lifetime. This corresponds to maximum pump energy transferred in the laser crystal of 167 mJ. The laser output energy as a function of the pump energy from the luminescent concentrator

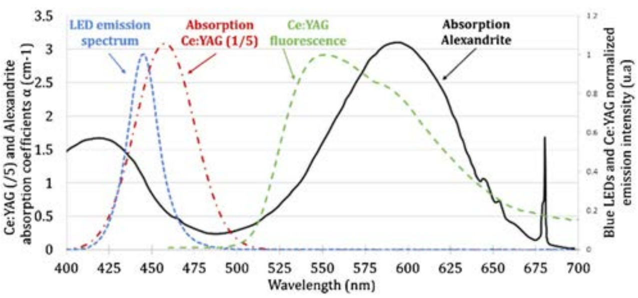

Fig. 1. Ce:YAG (red), alexandrite (black) absorption coefficients for light polarized parallel to the *b* axis. Emission spectra of Ce:YAG (green) and LED (blue) in the pulsed regime (260 μs, 5 A, 10 Hz, room temperature).

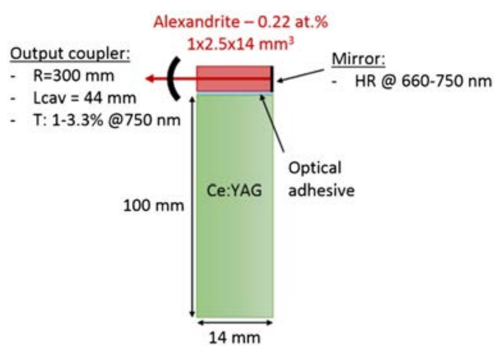

Fig. 2. Setup of the experiment. LEDs are located above and underneath the Ce:YAG concentrators.

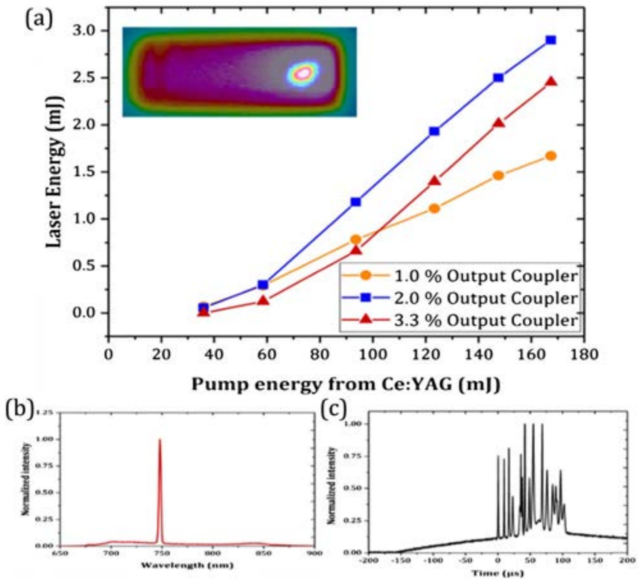

Fig. 3. (a) Laser output energy versus LC pumping energy for 1.0%, 2.0%, and 3.3% transmissions output couplers. Inset: spatial profile of the laser (overlaid with the fluorescence spatial profile of the 2.5 mm × 1 mm emitting facet of the alexandrite crystal that allows the position of the laser beam in the laser crystal to be visualized). (b) Laser spectrum in free running operation. (c) Laser and fluorescence temporal profile.

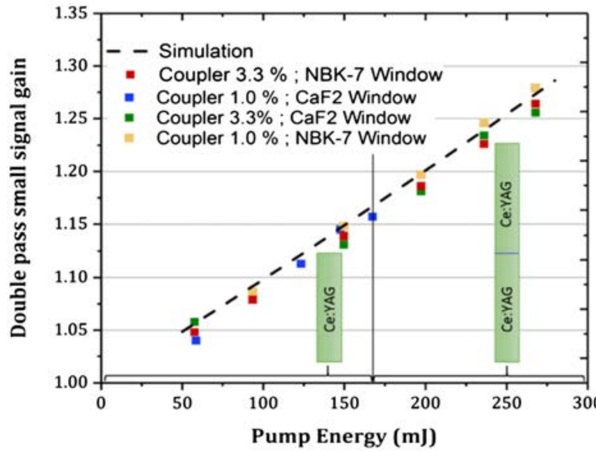

Fig. 4. Small signal gain per double-pass versus the pump energy: numerically estimated (dashed line) and measured for different output couplers and flat windows (dots).

is plotted in Fig. 3 for several output couplers. A maximum laser energy of 2.9 mJ at 748 nm is demonstrated with the 2.0-%-transmission output coupler [Figs. 3(a) and 3(b)]. The output beam is TEM00 with a diameter of 125 μm [Fig. 3(a) inset]. The laser operates during 108 μs with a buildup time of 152 μs [Fig. 3(c)]. This corresponds to a peak power of 26 W. For a pump energy of 167 mJ, the optical efficiency is then 1.7%. The total energy emitted by the 1120 LEDs is 990 mJ, which corresponds to a global efficiency of 0.29%. This work represents the first demonstration of LED-pumped Cr^{3+}-doped crystal. It is an important breakthrough for visible lasers using this technology as a step ahead compared with LED-pumped Nd^{3+}-doped gain media.

In order to have a full characterization of our system, we measured the small signal gain (per double pass) in the cavity. It is defined by $G_0 = P_{\text{out}}/P_{\text{in}}$ with P_{in} and P_{out} the power before and after one roundtrip in the crystal. This represents a key parameter in our way to laser amplification. In this study, we choose to investigate the evolution of the laser threshold as a function of intracavity losses [17]. Calibrated Fresnel losses are inserted in the cavity using a flat window with a variable incidence angle θ around Brewster angle. The laser being horizontally polarized, the losses per roundtrip in the cavity can be written as

$$R(\theta) = R_{pl} \cdot R_{O/c} \cdot \left[1 - \left(\frac{n_w \cdot \cos\theta - \cos\theta'}{n_w \cdot \cos\theta + \cos\theta'}\right)^2\right]^4,$$

$$\text{with} \quad \sin\theta' = \sin\theta / n_w, \qquad \textbf{(1)}$$

with R_{pl} being the contribution of passive losses, $R_{o/c}$ the reflectivity of the output coupler, n_w the refractive index of the flat window. The term in brackets represents the Fresnel reflections on a diopter. It is elevated to the power of 4 as the number of window interfaces encountered by the laser per round-trip in the cavity.

At threshold, small signal gain and losses are balanced. Then, G_0 is retrieved with the knowledge of θ, R_{pl}, $R_{o/c}$, and n_w. The passive losses were experimentally measured using the Findlay–Clay method [18] measuring the pump power at threshold for different output couplers. The passive losses are estimated at 2.2%. Measurements of the small signal gain are carried out with two different flat windows: the first one is an 8 mm thick N-BK7 glass plate with a refractive index of 1.51; the second is a 2 mm thick CaF_2 plate with a refractive index of 1.43. Two different output couplers with transmission of 1.0% and 3.3% are used (Fig. 4). The combination of materials having different refractive index and several output couplers provides better reliability of the study.

Figure 4 shows a double-pass small signal gain of 1.15 for a pump energy of 167 mJ launched in the alexandrite crystal. The small signal gain per double pass is also calculated numerically. These simulations require an estimation of the local pump density in the gain medium. This is performed using LightTools, a ray tracing software based on the Monte Carlo algorithm. The experimental results are similar to numerical simulations: the gain following a linear increase versus the energy. As no limitation occurs, we investigate power scaling to explore the potential of the setup.

Power scaling can be easily performed: the Ce:YAG luminescent concentrators length is doubled (200 mm) using two Ce:YAG concentrators bonded to each other (with a UV curing optical adhesive). This 200 mm long Ce:YAG concentrator can deliver up to 268 mJ to the gain medium. In this configuration, 2240 LEDs are used. The irradiance reached 8.5 kW/cm^2. The small signal gain still increases linearly and reaches 1.28 at maximum power.

Alexandrite can be seen, thanks to its tunability, as a good alternative to Ti:sapphire femtosecond amplifier. Indeed, several alexandrite-based femtosecond amplifiers have been developed during the 1990s [14,15]. Nevertheless, the weakness of these systems lies in the pump source. LED pumping then brings a revival of interest for alexandrite-based fs amplifiers.

We test the amplification performance of LED-pumped alexandrite with a CW Ti:sapphire laser injection. In this

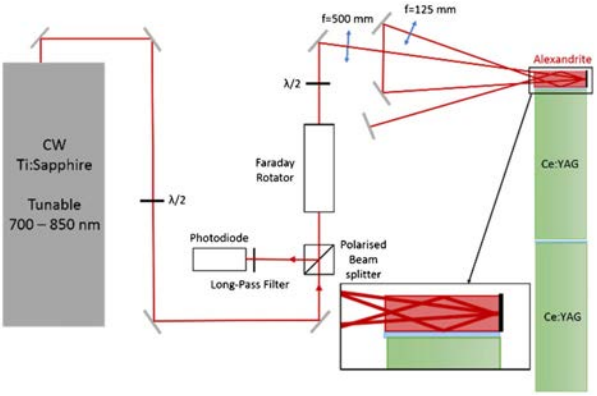

Fig. 5. Experimental setup of the tunable multipass amplifier for a CW Ti:sapphire laser.

experiment, the alexandrite dimensions are 1 mm × 1 mm × 14 mm to permit us to optimize the laser gain area. First, we design a setup that enables two double-passes in the 14 mm long alexandrite crystal using advantageously the possibility of total internal reflection in our crystal. The first double pass has a V-shape, and the second one has a rhombus shape. The number of passes through the crystal is then doubled using a Faraday rotator combined with a polarizer and a half-wave plate. It leads to eight single-passes into the alexandrite crystal with a good overlap of the gain zone (see the experimental setup in Fig. 5).

The signal is recorded using a photodetector and oscilloscope. Tuning the emission wavelength of the Ti:sapphire laser, the small signal gain after eight passes into the crystal can be plotted versus different wavelengths between 710 nm and 850 nm (Fig. 6). A maximum gain of 4 was measured at 750 nm using the 200 mm LC configuration and at the maximum pump energy (268 mJ). The small signal gain remains higher than 2 between 710 nm and 790 nm. The temporal profile of small signal gain in the alexandrite crystal is represented in the inset Fig. 6. This profile follows the dynamics of population inversion in the gain medium. When the Ti:sapphire wavelength is shifted closer to 700 nm reabsorption processes makes the gain collapse. Above 800 nm the gain is decreasing as expected from the alexandrite emission cross section.

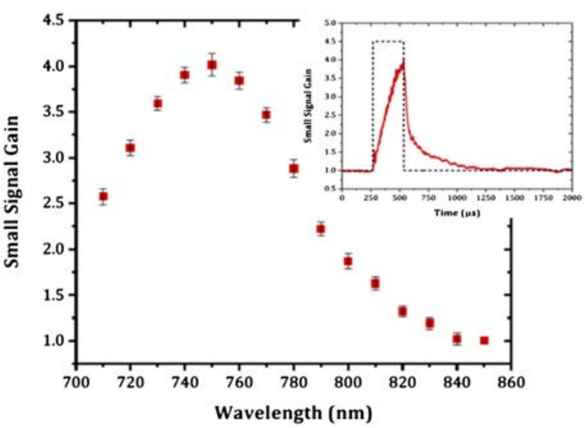

Fig. 6. Amplification of a CW Ti:sapphire laser as a function of the wavelength at maximum pumping. Inset: temporal profile of the small signal gain (red) LEDs activation profile (dotted black).

In conclusion, the first LED-pumped chromium-doped crystal laser oscillator and amplifier are presented. An alexandrite crystal is pumped by an innovative pump head [9] relying on blue LEDs and a Ce:YAG luminescent concentrator enhancing the LEDs radiance by a factor of 15. This pump source emits at 550 nm and benefits all of the advantages of LEDs. It permits us to deliver high radiance light in a spectral range where laser diodes show limited performances and are relatively expensive. A laser operating in free running at 10 Hz is demonstrated with output energies up to 2.9 mJ at 748 nm. The pumping system can be easily power scaled and with a pump energy of 268 mJ. In this case, the small signal gain is linear versus pump energy and reaches 1.28 at maximum. These results are in close agreement with numerical simulations. Driven by this interesting result, we performed small signal gain measurements in amplifier configuration. The amplifier design is an eight-pass multipass amplifier whose injection can be tunable using a CW Ti:sapphire laser. The maximum gain is 4 at the wavelength of 750 nm, and a tunability between 700 and 830 nm has been performed. The small signal gain remains higher than 2 between 700 nm and 790 nm. Nevertheless, for interesting perspectives, it is worth to note that gain performance above 800 nm could be improved by increasing the temperature of the crystal, as demonstrated in [15].

Based on this work, it is now possible to imagine innovative simple LED-based multipass amplifiers in the visible. This could result in the reduction of the price of current alexandrite CW systems and also Ti:sapphire femtosecond amplifier ideally with 1 and 2 orders of magnitude.

Funding. Agence Nationale de la Recherche (ANR) (ANR-12-BS09-0012-01).

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