Appl. Phys. B 89, 543–547 (2007)
DOI: 10.1007/s00340-007-2834-0

Applied Physics B
Lasers and Optics

Rapid communication

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Broad-band regenerative laser amplification in ytterbium-doped calcium fluoride (Yb:CaF_2)

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Received: 14 August 2007/Revised version: 26 September 2007
Published online: 3 November 2007 •

ABSTRACT An output pulse energy of 17.3 mJ has been achieved with a diode-pumped Yb:CaF_2 regenerative laser amplifier. The bandwidth of the output pulse spectrum was 7.3 nm, being seeded with femtosecond pulses stretched to 2.2 ns. In cw operation a tuning range of 80 nm has been observed. A maximum pulse energy of 44 mJ at a repetition rate of 1 Hz has been obtained in Q-switched mode. The laser damage threshold of a Yb:CaF_2 crystal has been determined at a wavelength of 1064 nm and a pulse duration of 10 ns.

PACS 42.55.Ah; 42.55.Xi; 42.70.Hj

1 Introduction

Recently, ytterbium-doped calcium fluoride (Yb:CaF_2) has attracted considerable interest for femtosecond pulse generation [1, 2] and widely tunable cw-laser operation [2, 3]. Although a well-known host material [4, 5], CaF_2 doped with rare-earth ions has become increasingly attractive as diode-pumped laser material for a number of reasons [6]. It can be easily grown by the Czochralski, Bridgman, or temperature-gradient techniques [7]. Undoped single-crystalline CaF_2 with optical quality is readily available with diameters up to 380 mm for lithographic applications [8–10]. Meanwhile, the growth of single-crystalline samples of Yb:CaF_2 with lateral dimensions of 76 mm has been proven [7].

The substitution of trivalent ytterbium ions for Ca^{2+} and thus the necessary charge compensation leads to a rich multi-site structure including isolated ions up to clusters [11–14] and results in broad absorption and emission bands. Furthermore, in comparison to various ytterbium-doped laser materials such as garnets, oxides, silicates, vanadates, or tungstates [15], Yb:CaF_2 comprises a long fluorescence lifetime of 2.05 ms [16], which reduces the number of required pumping diodes in order to accumulate a certain amount of optical energy within the gain medium of a pulsed laser amplifier. Co-doping with Na^+ prevents the formation of Yb^{2+} ions [17–19], supports the homogeneous distribution of the Yb^{3+} ions, and thus improves the quantum efficiency by reduced quenching effects [20].

In addition, CaF_2 with a thermal conductivity of 9.7 $W\,m^{-1}\,K^{-1}$ is well suited for laser systems with high repetition rates and thus high average output power [1, 21]. However, the reduction of the thermal conductivity with increasing Yb^{3+} concentration has to be taken into account [22]. For example, at an Yb concentration of 5 mol. % the thermal conductivity decreases to 5.0 $W\,m^{-1}\,K^{-1}$ [23]. In general, alkaline-earth fluorides show a wide optical transmission range, a low dispersion behavior at small linear and nonlinear refractive indices, and thus limited nonlinear effects under intense laser irradiation.

In this paper, we present to our knowledge the first broad-band regenerative amplification of femtosecond pulses to the mJ level within single-crystalline Yb^{3+}:CaF_2. As an important application of this new laser material, Yb:CaF_2 might be attractive for diode-pumped chirped pulse amplifier (CPA) systems. A fundamental milestone in the development of a certain gain medium towards a favorable laser material for ultra-short-pulse laser systems is the proof of the bandwidth at a high gain in a laser amplifier.

2 Crystal parameters

For this work a cylindrical Yb^{3+}:CaF_2 crystal with a diameter of 15 mm and a length of 35 mm has been grown by the Czochralski technique with rf heating at the Institute for Crystal Growth (IKZ), Berlin, Germany. Following the binary phase diagram of CaF_2–YbF_3 [24], the segregation coefficient of YbF_3 in CaF_2 is a little below one. Therefore, the dopant concentration of the starting material had to be chosen somewhat higher than needed in the crystal. Thus, in accordance with the phase diagram, a slight segregation takes place during crystal growth. Starting with an YbF_3 content of 3.5 mol. % in the melt, the upper part of the crystal contained 3.28 ± 0.01 mol. % YbF_3 and the lower part 3.35 ± 0.01 mol. % YbF_3 (both values measured by inductively coupled plasma-optical emission spectroscopy (ICP-OES)). However, due to the flat crystal–melt interface of the growing crystal the dopant is homogeneously implemented along

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the crystal cross section. In order to reach a single-pass absorption of 64% at 940 nm (87% when applied at Brewster's angle), a sample crystal was cut to a length of 7.6 mm. The surfaces (parallel to the 111 direction) of the crystal samples were finished by pitch polishing (Hellma Optik GmbH, Germany) with diamond polishing compounds (grain size 0.1 μm).

Prior to the laser experiments the optical transmission properties such as wave-front distortion, striae, and stress birefringence were investigated. Striae and inhomogeneities of the refractive index can be detected with a striae analyzer filtering the low diffraction orders at the Fourier plane (Fig. 1a). Figure 1b illustrates the white-light transmission through the crystal between crossed polarizers, which gives information about intrinsic stress between the grain boundaries of the crystal. A tolerable polarization rotation of a maximum of 1.7° at single-pass transmission has been measured. The grain boundaries close to the crystal surface can be visualized by X-ray diffraction within a small acceptance angle (here 0.2°) shown in Fig. 1d. Applying a phase-retrieval algorithm a double-pass wave-front distortion of 0.42 λ around the crystal core has been determined with a Michelson interferometer at a wavelength of 633 nm (Fig. 1c). Considering the recency and the current state of developing Yb-doped calcium fluoride, the investigated crystal shows a high grade of optical quality.

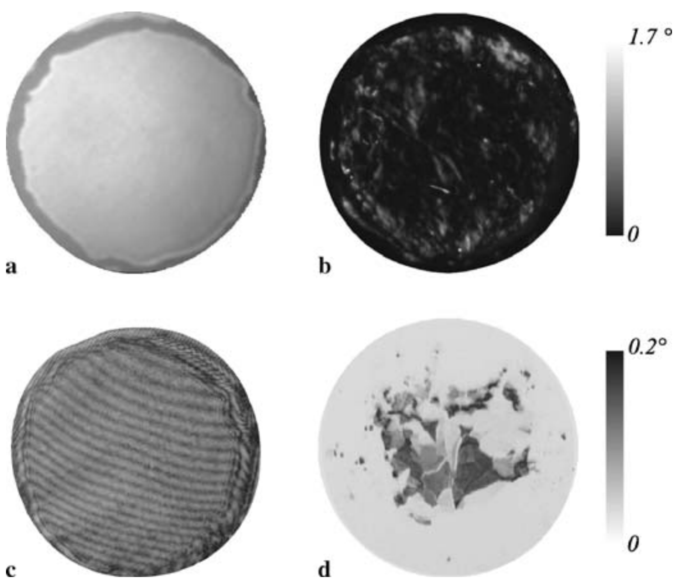

FIGURE 1 Optical properties of the Yb^{3+}:CaF_2 crystal: (**a**) striae analysis, (**b**) polarimetry, visualization of intrinsic stress birefringence, (**c**) Michelson interferometry, analysis of the wave-front distortion ($\lambda = 633$ nm), (**d**) X-ray topography, visualization of grain boundaries at the crystal surface

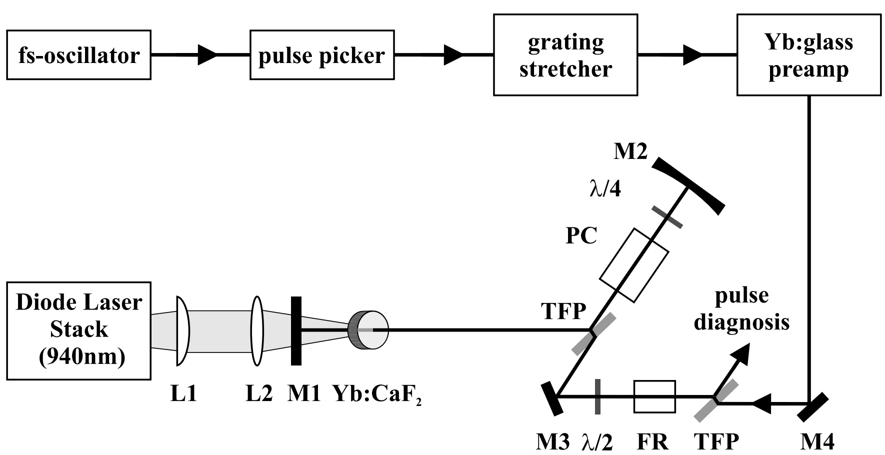

FIGURE 2 Experimental setup: L1, cylindrical lens ($f = 115$ mm); L2, spherical lens ($f = 75$ mm); M1, dichroic flat mirror (HR 1020–1060 nm, AR 930–950 nm); M2, concave mirror (radius of curvature 3000 mm); TFP, thin-film polarizer; λ/4, quarter-wave plate; PC, Pockels cell (DKDP); M3 and M4, plane HR deflecting mirrors; λ/2, half-wave plate; FR, Faraday rotator

3 Tunable cw-laser operation

Figure 2 shows the setup of the laser system. The gain medium (Yb:CaF_2) was pumped by a pulse-driven stack of 25 fast axis collimated diode-laser bars (Jenoptik Laserdiode GmbH, Germany), having a peak output power of 2.5 kW. For re-collimation of the slow axis the diode stack was followed by a cylindrical lens ($f = 115$ mm). The pump radiation was focused into the gain medium by a spherical lens ($f = 75$ mm) yielding a peak intensity of 83 kW cm^{-2} at an elliptical pump spot (1.4 mm × 1.9 mm (FWHM)) with a Gaussian intensity distribution. Following the specifications of the pumping diodes the maximum pump duration was limited to 1.5 ms at a repetition rate of 1 Hz. The transversally cooled Yb:CaF_2 crystal was mounted in a water-cooled brass assembly with indium foil in between, whereas the polished surfaces were orientated at Brewster's angle to the expected laser mode. At full pump power a polarization contrast of 1 : 400 due to thermally induced stress birefringence has been determined at single-pass transmission.

A plane dichroic mirror (M1) acting as a wavelength coupler and a concave mirror (M2) with 3 m radius of curvature represent a hemispherical resonator (substrates and coatings provided by Layertec GmbH, Germany). Due to negative thermal lensing [21] of the colinearly end-pumped Yb:CaF_2 crystal, the cavity length was 95 cm for optimum stability.

Prior to the pulse-amplification experiments the tuning range of Yb:CaF_2 with an intra-cavity SF10 prism has been investigated. Corresponding to the broad emission spectrum of Yb:CaF_2 a tuning range of 80 nm (see Fig. 3a) has been observed at maximum pump power. In the case of quasi-cw operation (pulse duration: 1 ms) a maximum average output power of 340 mW without the intra-cavity prism at a repetition rate of 1 Hz has been obtained. Applying the moving knife-edge method a beam quality (M^2) of 1.21 was found. In spatially multi-mode operation where the match-

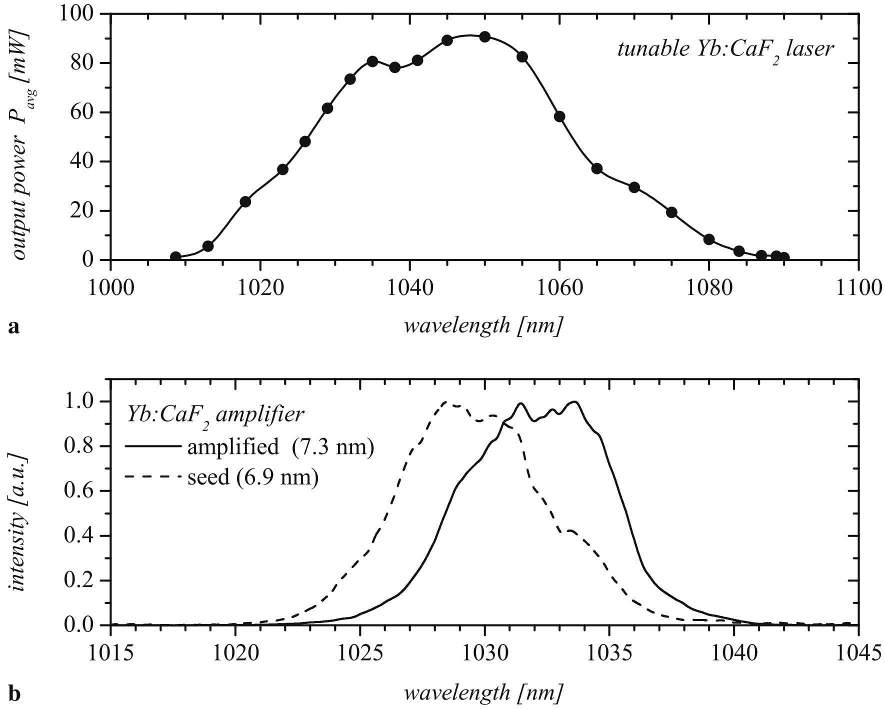

FIGURE 3 (**a**) Average output power (duty cycle: 0.1%) vs. center wavelength: tuning curve of Yb:CaF$_2$ laser in quasi-cw operation with SF10 prism inside the optical resonator; (**b**) spectra of seed ($\lambda_0 = 1030$ nm) and amplified ($\lambda_0 = 1032.5$ nm) laser pulses, chirped pulse amplification (CPA), pump duration: 1.5 ms, pump power: 1.3 kW, number of round trips: 32, total gain: 1.4×10^3

ing of the laser mode and the elliptical pump profile is increased, the average output power of 340 mW is achieved when pumping at 1.1 W (950 mW absorbed pump power).

4 Amplification and *Q*-switching experiments

For *Q*-switched pulse generation and regenerative amplification a thin-film polarizer (TPF), a quarter-wave plate, and a Pockels cell were placed in the resonator. A DKDP Pockels cell (QX 1020, Cleveland Crystals Inc., USA) with 1 cm clear aperture has been used. The Pockels cell driver allows for fast switching with rise and fall times (10%–90%) of 2.6 ns (both driver and delay generator provided by BME-Bergmann KG, Murnau, Germany). In *Q*-switched operation the laser generated pulses by cavity dumping with a pulse duration down to 6.4 ns (FWHM) and a pulse energy up to 44 mJ before optical damage to the coating of mirror M1 occurred. At a pump power of 1.3 kW a build-up time of 540 ns is required in order to achieve maximum output pulse energy. The pulse width was recorded with an InGaAs photodiode (ET-3000, EOT Inc., USA) having a cutoff frequency of > 2 GHz followed by a 500-MHz digital sampling oscilloscope.

The seed pulses with a pulse width of 85 fs were generated in a commercial Ti:sapphire oscillator (Mira 900, Coherent Inc., USA) which was tuned to a center wavelength of 1030 nm. After electro-optical pulse selection, pulses with a pulse energy of 1.3 nJ and a bandwidth of 18 nm were stretched to 2.2 ns by a four-pass stretcher (General Atomics Inc., USA). The stretcher with a hard-clip bandwidth of 32 nm incorporates a 14-in gold grating with 1400 lines per mm. At a repetition rate of 1 Hz the pulses were then amplified to the 10 µJ level by a diode-pumped regenerative Yb:glass [25, 26] pre-amplifier analogous to the described Yb:CaF$_2$ system. In order to separate input and output pulses a half-wave plate, a thin-film polarizer (TFP), and a Faraday rotator were placed between the amplifiers. Both input and output spectra have been determined with a fiber optic spectrometer (USB 2000, Ocean Optics Inc., USA) with a spectral resolution of 0.4 nm (see Fig. 3b). Major gain narrowing of the pulse spectrum is due to the Yb:glass pre-amplifier. Furthermore, corresponding to the gain spectrum of Yb:CaF$_2$, the input spectrum (center wavelength: 1030 nm) was red shifted to 1032.5 nm. A maximum pulse energy of 17.3 mJ at 32 round trips was determined. In order to prevent laser-induced damage the output pulse energy was not increased to a higher level, whereas the gain of the amplifying medium remained unsaturated.

In a next step a direct amplification of the un-stretched oscillator pulses has been investigated. Without the stretcher and pre-amplifier the center wavelength could be slightly tuned and the full oscillator bandwidth of 18 nm has been available. The fs pulses at the nJ level have been amplified to 50 µJ without optical damage. Owing to the intracavity dispersion (mostly the DKDP crystal of the Pockels cell) the pulses were stretched to 550 fs, which has been measured by a second-order autocorrelation. When seeded at a center wavelength of 1045 nm an output bandwidth of 13.4 nm at a total gain of 4×10^4 has been achieved (see Fig. 4). Both the bandwidth and the shift of the center wavelength are plotted vs. the seed center wavelength in Fig. 5. During the experiment the input and output pulse energies were constant. As a result the red shift of the output center wavelength as well as the bandwidth increase at lower wavelengths. The achieved total gain as well as the single-pass gain are similar to those values observed at regenerative amplification in Yb-doped laser glass (Yb:glass pre-amplifier, see Fig. 1). Compared to many other Yb-doped host materials (e.g. Yb-doped garnets or tungstates) Yb:CaF$_2$ exhibits a low emission cross section at the laser wavelength, which in general complicates efficient pulse energy scaling. Thus, the presented amplification experiment can be considered to be a proof of principle in order to apply Yb:CaF$_2$ as gain medium in diode-pumped femtosecond CPA systems.

Finally, in order to estimate the suitability of Yb:CaF$_2$ for high-power laser systems the optical damage threshold of the gain medium has been determined. According to ISO 11254-1 1-on-1 measurement [27], we have measured the damage threshold in single-shot tests with a beam diameter of 30 µm. A *Q*-switched Nd:YAG laser with 1.2 J

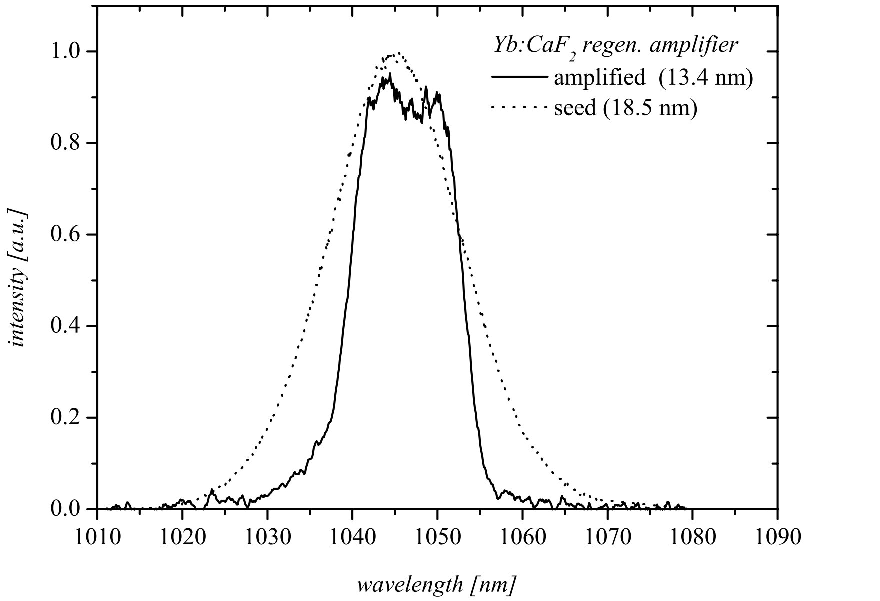

FIGURE 4 Spectra of seed ($\lambda_0 = 1045$ nm) and amplified ($\lambda_0 = 1046$ nm) laser pulses, direct amplification of oscillator pulses, pump duration: 1.5 ms, pump power: 1.3 kW, number of round trips: 49, total gain: 4×10^4

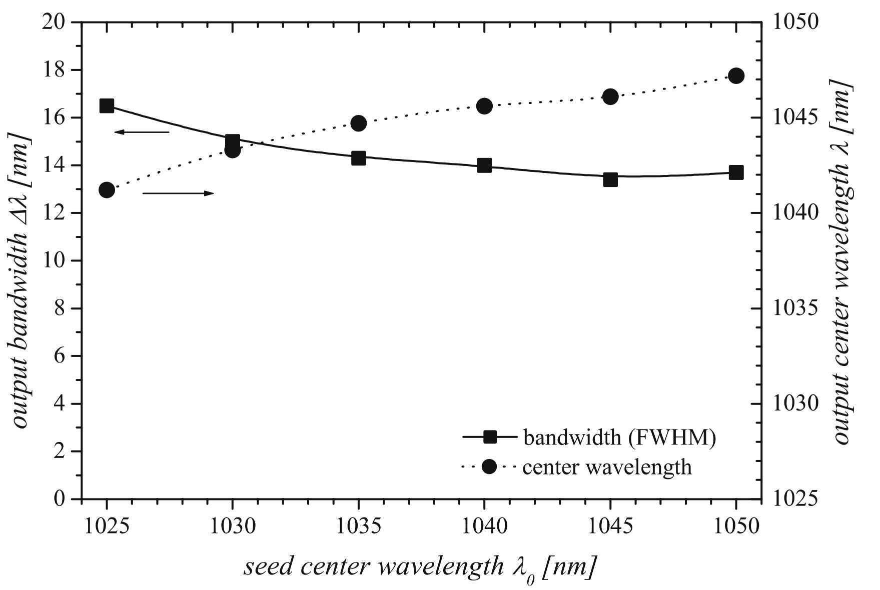

FIGURE 5 Bandwidth (FWHM) and center wavelength vs. seed center wavelength, direct amplification of oscillator pulses, seed bandwidth: 18 nm, output pulse energy: 50 μJ, total gain: 4×10^4

maximum output pulse energy (Powerlite Precision II 8000, Continuum Inc., USA) was followed by an attenuator and a focusing lens with a focal length of 50 cm. During our investigation we have defined the damage threshold F_{thr} as the fluence where damage barely occurs. At a pulse duration of 10 ns (FWHM) and a center wavelength of 1064 nm the surface damage threshold of fused silica (139 J cm^{-2}) and different ytterbium-doped laser materials such as Yb:YAG (16 J cm^{-2}), Yb:KGW (23 J cm^{-2}), and Yb:fluoride–phosphate glass (32 J cm^{-2}) has been measured for comparison. In the case of Yb:CaF_2 a damage threshold of 52 J cm^{-2} at the same laser parameters has been found.

5 Conclusion

We have shown the amplification of chirped fs pulses within a regenerative Yb:CaF_2 amplifier at the mJ level. When seeded with pulses at the 10 μJ level an output pulse energy of 17.3 mJ and a bandwidth of 7.3 nm (FWHM) have been achieved. The bandwidth of the output pulses allows us to re-compress the pulses down to 215 fs. In the case of seeding with unchirped fs pulses a bandwidth of up to 16 nm (FWHM) at a high gain up to the 50 μJ level has been observed. Thus, the potential of Yb:CaF_2 for diode-pumped fs-pulse amplification was successfully demonstrated. Furthermore, Yb:CaF_2 shows a high damage threshold for ns pulses, which gives an optimistic perspective towards being a promising gain medium in diode-pumped lasers for high pulse energies.

ACKNOWLEDGEMENTS This work has been supported by the German Federal Ministry of Education and Research (BMBF) under Contract No. 03ZIK052 (ultra optics). We thank M. Rabe for crystal growth experiments and I. Uschmann, O. Wehrhan, H. Marschner, and B. Lüdge for the X-ray diffraction and interferometric measurements.

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