Apatite-structure crystal, $\text{Yb}^{3+}:\text{SrY}_4(\text{SiO}_4)_3\text{O}$, for the development of diode-pumped femtosecond lasers

F. Druon, S. Chénais, P. Raybaut, F. Balembois, and P. Georges

Laboratoire Charles Fabry de l'Institut d'Optique—Ecole Supérieure d'Optique, Unité Mixte de Recherche 8051 du Centre National de la Recherche Scientifique, Centre Universitaire d'Orsay, Bâtiment 503, 91403 Orsay, France

R. Gaumé, P. H. Haumesser, B. Viana, and D. Vivien

Laboratoire de Chimie Appliquée de l'État Solide, Unité Mixte de Recherche 7574 du Centre National de la Recherche Scientifique, École Nationale Supérieure de Chimie Paris, France

S. Dhellemmes, V. Ortiz, and C. Larat

THALES Research and Technology, Domaine de Corbeville, 91404 Orsay Cedex, France

Received July 1, 2002

We report what is believed to be the first diode-pumped $\text{Yb}^{3+}:\text{SrY}_4(\text{SiO}_4)_3\text{O}$ laser. We investigate both the cw and the mode-locked regimes. In the cw regime an average output power of 1.05 W is demonstrated for 3.6 W of incident pump power. In the femtosecond regime a pulse duration of 94 fs for an average power of 110 mW is obtained at a central wavelength of 1068 nm. This is, to our knowledge, the first femtosecond mode-locked oscillator made with an Yb-doped apatitelike crystal. These properties are related to the unique spectroscopic properties of Yb^{3+} ions in this atypical apatite-family crystal. © 2002 Optical Society of America
OCIS codes: 140.5680, 140.3480, 140.4050, 140.3070.

In the development of diode-pumped femtosecond lasers, Yb-doped crystals have attracted strong interest because of their usefulness in simplifying, compacting, and reducing the cost of ultrashort-pulse solid-state lasers.^{1–6} Yb^{3+} -doped materials have the advantage over other materials of a simpler electronic structure based on only two manifolds, which is attractive for laser operation because of the absence of deleterious effects such as excited-state absorption, upconversion, and luminescence self-quenching. The drawback of this simple electronic structure is a quasi-three-level laser scheme that requires relatively high-brightness pumping. However, Yb-doped materials also combine the capacity to be pumped at 980 nm by bright high-power InGaAs diodes, a very low quantum defect (<10%), and the possibility of being highly doped without quenching by the cross-relaxation process, a common phenomenon for Nd-doped materials. These advantages can then lead to compact, simple, and efficient laser systems. Moreover, an important advantage of Yb-doped materials over their Nd-doped counterparts is their broad emission bandwidth, which is crucial for the generation of ultrashort pulses.

From the point of view of developing both highly efficient and ultrashort-pulse laser sources, the investigation of new crystal hosts for Yb has also sparked a great deal of interest. Yb:apatite-class crystals, particularly fluoroapatites, are interesting in that they have very intense emission cross-section peaks, up to $6.2 \times 10^{-20} \text{ cm}^{-2}$.^{7–9} In contrast, the spectral bandwidths of these peaks are very narrow (4 nm), which makes them incompatible with femtosecond-pulse generation.¹⁰ Investigating new Yb:silicate

apatite crystals with a broader emission band is the topic of current work. These crystals must have high structural disorder in their lattice. The silicate oxyapatite crystal $\text{Yb}:\text{SrY}_4(\text{SiO}_4)_3\text{O}$ (Yb:SYS) presents a broad emission band because of an equally random distribution of Sr and Y on the same Wyckoff site, namely, 4f, see Fig. 1.^{11–13} Moreover, the Yb ions that statistically substitute Y (because of the compatibility of charge and ionic radii) also have a second site for substitution, 6h (Fig. 1). The ratio of substitution between these two sites is 3:1 at the advantage of 6h. Because of this multisite occupancy on the Y sites and cationic disorder, Yb:SYS exhibits very broad emission and absorption bands, as shown in Fig. 2. The peak emission cross section decreases drastically compared with those of other apatite crystals, with a value of only $0.44 \times 10^{-20} \text{ cm}^2$ at 1040 nm (in comparison

Fig. 1. Representation of the Yb:SYS structure: The Wyckoff (4f) site can be occupied randomly by Sr and Y (and occasionally Yb when doped); 6h sites, by Y (and occasionally Yb when doped).

Fig. 2. Emission and absorption cross-section curves of Yb:SYS. Inset, Yb:SYS emission cross-section curve compared with those of other ultrabroadband Yb-doped materials.¹¹

with $6.2 \times 10^{-20} \text{ cm}^2$ at 1046 nm in the Yb:Sr₅(PO₄)₃ (Yb:SFAP); however, the emission bandwidth indeed increases compared with those of other apatite crystals and is $\sim 73 \text{ nm}$. Compared with other very broadband Yb-doped materials (e.g., Yb:glass, Yb:Ca₄GdO(BO₃)₃ (Yb:GdCOB), or Yb:Sr₃Y(BO₃)₃ (Yb:BOYS), Yb:SYS crystal presents a broader and more intense emission peak (Fig. 2), with a relatively high emission cross section of almost 1100 nm. The thermal properties of the SrY₄(SiO₄)₃O (SYS) crystals are also relatively good: The thermal conductivity of undoped SYS is $2.85 \text{ Wm}^{-1} \text{ K}^{-1}$ along the $\tilde{c}$ axis and $1.6 \text{ Wm}^{-1} \text{ K}^{-1}$ for the axis perpendicular to $\tilde{c}$. Furthermore, it is known that thermal conductivity may decrease with the doping level: For Yb:SYS (5.5% doped), the thermal conductivity is $2.85 \text{ Wm}^{-1} \text{ K}^{-1}$ ($\parallel \tilde{c}$) and $1.31 \text{ Wm}^{-1} \text{ K}^{-1}$ ($\perp \tilde{c}$). In addition, the Yb:SYS compound could be relatively easily grown by use of the Czochralski method despite the compound's melting point of $\sim 1900^\circ \text{C}$. Accordingly, the optical quality obtained for a 5.5%-Yb-doped 21-mm-diameter boule was excellent. Single-crystalline pieces are shown in Fig. 3. Finally, Yb:SYS has the advantage of a large ground-state crystal-field total splitting of 810 cm^{-1} ,¹¹ which is important for reducing the effect of thermal population in this quasi-three-level laser material. In this Letter we present what is to our knowledge the first demonstration of a diode-pumped Yb:SYS laser. This laser was studied in both the cw and the femtosecond regimes.

For the cw regime the experiment was performed with a 5.5%-doped ($8.4 \times 10^{20} \text{ Yb}^{3+} \text{ atoms/cm}^3$) 5-mm-long Yb:SYS crystal. The pump source was a $1 \mu\text{m} \times 100 \mu\text{m}^2$ active-area laser diode emitting 4 W of cw power at 975 nm. The incident power on the crystal was 3.6 W, and the crystal absorbed 3.3 W. The pump beam was reshaped by a cylindrical telescope (along the slow axis of the diode's active area) and was focused onto the crystal to a spot size of $170 \mu\text{m} \times 30 \mu\text{m}$. The cavity design was a standard V-shaped cavity made from three mirrors: a dichroic mirror (highly reflecting at 1060 nm, highly transmissive at 980 nm) with a radius of curvature

of 50 mm, a 300-mm radius-of-curvature mirror, and a plane output coupler with a transmission of 10%. With such a cavity, the laser beam waist in the crystal was $160 \mu\text{m}$ in diameter. Under these conditions, we obtained up to 1.05 W of cw power at 1070 nm, which corresponds to an optical-optical efficiency of 32% (output power versus absorbed pump power). The beam profile is TEM₀₀, with an M^2 factor of 1.05. The laser threshold was obtained for an absorbed pump power of 1.4 W. By inserting a prism into the cavity, we also achieved tuning of the laser emission from 1020 to 1095 nm, with a FWHM bandwidth of 53 nm.

To reduce the reabsorption effect, for the femtosecond oscillator we performed the experiment with a shorter Yb:SYS crystal. We used a 5.5%-Yb-doped 3-mm-long Yb:SYS crystal pumped with a single-stripe $1 \mu\text{m} \times 100 \mu\text{m}$ diode. The crystal absorbed 1.6 W of the pump power. To compensate for the group-velocity dispersion a pair of SF10 prisms, separated by 30 cm was used (Fig. 4). To initiate mode locking in the laser we used a semiconductor saturable-absorber mirror (SESAM).¹⁴ The SESAM was composed of a Bragg mirror centered at 1060 nm and a low-temperature-grown 30-nm-thick InGaAs saturable-absorber single layer. To optimize the pulse fluence on this SESAM we focused the cavity mode to a

Fig. 3. Several pieces of 5.5%-doped Yb:SYS, along with a 1 Euro coin for scale.

Fig. 4. Experimental setup of the Yb:SYS mode-locked oscillator.

Fig. 5. (a) Autocorrelation and (b) spectrum of 94-fs pulses produced by the Yb:SYS laser.

spot-size diameter of 80 μm . The advantage of using a SESAM compared with Kerr-lens mode locking is that one does not have to perform critical alignment, which leads to a more-stable laser.¹⁴ Moreover, mode-locking instability is a critical point to be considered,¹⁵ especially in the case of Yb-doped materials, which move easily toward *Q*-switched regimes because of their long excited-state lifetimes (0.8 ms for Yb:SYS). In these conditions the shorter pulses obtained had a duration of 94 fs, assuming a sech^2 soliton shape for the autocorrelation (Fig. 5). The corresponding spectrum was centered at 1070 nm, with a FWHM bandwidth of 14 nm, which corresponds to a time-bandwidth product of 0.35. The average output power was 110 mW, and the repetition rate was 108 MHz (for comparison, the average output power when the SESAM was replaced with a standard highly reflecting mirror was 300 mW). We also obtained 130 mW of average power with 110-fs pulses at 1068 nm. In both cases the beam profile was TEM_{00} . At this point we were not able to reach shorter wavelengths or produce shorter pulses because of stability problems in the mode-locking process: The mode-locked regime with a single pulse per round trip became unstable. Indeed, when we tried to obtain shorter pulses by adjustment of the dispersion in the cavity or to reach shorter wavelengths by use of the slit, we observed, depending on the nature of the SESAM (temperature of 600 or 700 °C during the annealing process), the emergence of a cw component or double-pulse behavior.

In conclusion, we demonstrate, for the first time to our knowledge, diode-pumped cw and femtosecond oscillators based on a new Yb-doped apatite-type crystal. This silicate apatite crystal has the unique property of high structural disorder and thus exhibits a very broad emission band of 73 nm. Yb:SYS lasers are also particularly interesting because of their relatively high emission cross sections (compared with those of other very broadband Yb-doped materials), especially in the long-wavelength range (>1050 nm). We tested Yb:SYS crystal in a diode-pumped laser configuration in both the cw and femtosecond regimes.

In the cw regime we obtained as much as 1.05 W of output power at 1071 nm, with an incident power of 3.6 W. Laser operation was also observed over a wide wavelength range (75 nm) and up to 1095 nm. In the femtosecond regime, sub-100-fs pulses were generated near 1070 nm with an average power of 110 mW. This new Yb-doped apatite crystal seems to be a promising material for the development of ultrashort diode-pumped solid-state lasers.

F. Druon's e-mail address is frederic.druon@iota.u-psud.fr.

References

1. M. E. Fermann, A. Galvanauskas, G. Sucha, and D. Harter, *Appl. Phys. B* **65**, 259 (1997).
2. C. Hönninger, R. Paschotta, M. Graf, F. Morier-Genoud, G. Zhang, M. Moser, S. Biswal, J. Nees, A. Braun, G. Mourou, I. Johannsen, A. Giesen, W. Seeber, and U. Keller, *Appl. Phys. B* **69**, 3 (1999).
3. F. Druon, F. Balembois, P. Georges, A. Brun, A. Courjaud, C. Hönninger, F. Salin, A. Aron, F. Mougél, G. Aka, and D. Vivien, *Opt. Lett.* **25**, 423 (2000).
4. F. Brunner, G. J. Spühler, J. Aus der Au, L. Krainer, F. Morier-Genoud, R. Paschotta, N. Lichtenstein, S. Weiss, C. Harder, A. A. Lagatsky, A. Abdolvand, N. V. Kuleskov, and U. Keller, *Opt. Lett.* **25**, 1119 (2000).
5. H.-H. Liu, J. Nees, and G. Mourou, *Opt. Lett.* **26**, 1723 (2001).
6. F. Druon, S. Chénais, P. Raybaut, F. Balembois, P. Georges, R. Gaumé, G. Aka, B. Viana, S. Mohr, and D. Kopf, *Opt. Lett.* **27**, 197 (2002).
7. L. D. DeLoach, S. A. Payne, L. L. Chase, L. K. Smith, W. L. Kway, and W. F. Krupke, *IEEE J. Quantum Electron.* **24**, 1179 (1993).
8. S. A. Payne, L. D. DeLoach, L. K. Smith, W. L. Kway, J. B. Tassano, W. F. Krupke, B. H. T. Chai, and G. Louts, *J. Appl. Phys.* **76**, 497 (1994).
9. K. I. Schaffers, A. J. Bayramian, C. D. Marshall, J. B. Tassano, and S. A. Payne, in *Advanced Solid State Lasers*, C. R. Pollock and W. Bosenberg, eds., Vol. 10 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 1997), pp. 420–424.
10. X. Long, Y. P. Tong, P. M. W. French, and J. R. Taylor, *Opt. Commun.* **141**, 162 (1997).
11. P.-H. Haumesser, R. Gaumé, B. Viana, E. Antic-Fidancev, and D. Vivien, *J. Phys. Condens. Matter* **13**, 5427 (2001).
12. P.-H. Haumesser, R. Gaumé, B. Viana, G. Aka, D. Vivien, and B. Ferrand, *Advanced Solid State Lasers*, Ch. Marshall, ed., Vol. 50 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 2001), pp. 425–428.
13. P.-H. Haumesser, R. Gaumé, J.-M. Benitez, B. Viana, B. Ferrand, G. Aka, and D. Vivien, *J. Crystal Growth* **233**, 233 (2001).
14. U. Keller, *Appl. Phys. B* **58**, 347 (1994).
15. C. Hönninger, R. Paschotta, F. Morier-Genoud, M. Moser, and U. Keller, *J. Opt. Soc. Am. B* **16**, 46 (1999).