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Efficient lasing in diode-pumped Yb^{3+} : $CaF_2 - SrF_2$ solid-solution single crystals

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Abstract. **Single crystals of solid solutions of a high optical quality are grown in the concentration vicinity of the saddle point of the ternary $CaF_2 - SrF_2 - YbF_3$ system. Efficient lasing with a small Stokes shift (at 1025 nm) was obtained in 980-nm diode-pumped single crystals. The total lasing efficiency (with respect to the absorbed average pump power) was 59 % and the slope efficiency was 83 %.**

Keywords: *calcium fluoride, strontium fluoride, ytterbium fluoride, single crystals, diode pumping, lasing.*

As shown in papers [1, 2], ytterbium-doped CaF_2 single crystals are of great interest for the development of efficient tunable lasers and lasers emitting femtosecond pulses. The outlook for using ytterbium-doped fluoride single crystals to create near-IR lasers was demonstrated as early as 1993 in paper [3]. It was pointed out that an advantage of fluoride single crystals is the long excited-state lifetime. In addition, the presence of different ytterbium centres and their clusters leads to a considerable inhomogeneous broadening of absorption and luminescence lines, thereby providing conditions for the development of tunable and femtosecond lasers.

However, Yb^{3+} : CaF_2 single crystals known at present have a number of technological and spectral disadvantages, which can restrict their applications.

One of the technological disadvantages is a comparatively complex synthesis of single crystals because their quality is highly sensitive to variations in the synthesis parameters. Deviations from optimal parameters result in the formation of a cellular structure of grown single crystals, which makes them inadequate for laser applications (Fig. 1). In addition, as known from studies on doping fluoride single crystals with neodymium ions, cluster optical centres are formed in CaF_2 crystals at lower concentrations than, for example, in SrF_2 crystals. For this reason, single crystals of Yb^{3+} : $CaF_2 - SrF_2$ solid solutions were proposed instead of the known Yb^{3+} : CaF_2 crystals. It was shown in [4, 5] that such a ternary system has the so-called saddle point in which the compositions of the melt and crystal are the same, i.e. congruent melting takes place (Fig. 2). As a result, the stability of synthesis in the concentration vicinity of this

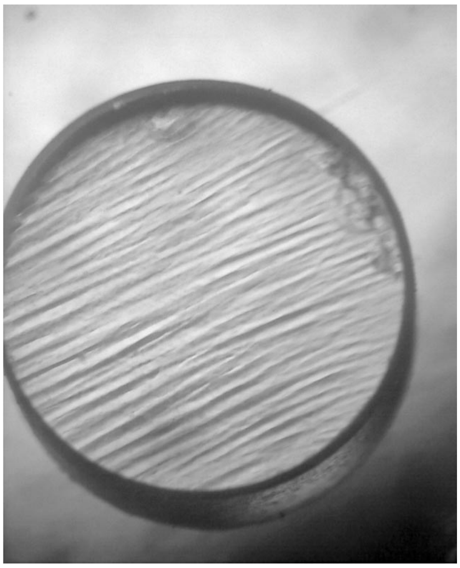

Figure 1. Cellular structure of a CaF_2 : Yb^{3+} crystal.

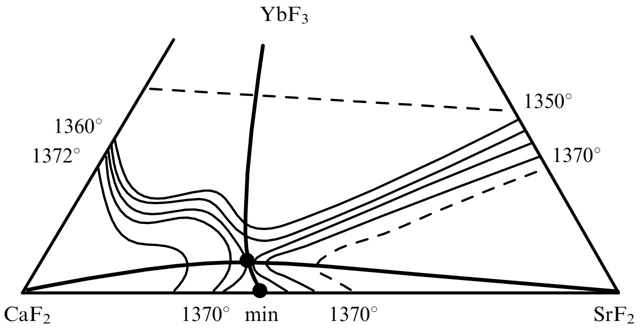

Figure 2. Isotherms (°C) of the liquidus surface in the concentration triangle of the $CaF_2 - SrF_2 - YbF_3$ system. The heavy lines are the separatrices of crystallisation lines. Shown are the saddle point with coordinates 60 ± 2 % CaF_2, 34 ± 2 % SrF_2, 6 ± 2 % YbF_3 ($T = 1370 \pm 5$ °C) and the minimum point with coordinates 60 % CaF_2 – 40 % SrF_2 ($T = 1362$ °C). The dashed curve is the boundary of the region of existence of the $(Ca_{1-y}Sr_y)Yb_xF_{2+x}$ fluorite solid solution.

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point is considerably higher, which makes it possible to obtain high-quality single crystals of solid solutions without cells at molar concentrations of Yb^{3+} ions equal to 3 % – 10 %.

We studied the spectral and lasing properties of ytterbium-doped fluoride materials. Samples were 1 – 2-mm-thick plates made of $Yb^{3+}:CaF_2$ and $Yb^{3+}:SrF_2$ single crystals and $Yb^{3+}:CaF_2-SrF_2$ solid solutions with the molar concentration of ytterbium ions from 4 % to 6 % to provide approximately 80 % absorption at the pump wavelength. The luminescence spectra and luminescence kinetics of samples were studied upon excitation by a tunable nanosecond $F_2^+:LiF$ colour centre laser. Luminescence spectra were recorded with an MDR-2 monochromator equipped with a FEU-83 photomultiplier and a TDS 3032 digital oscilloscope. A digitised signal was recorded in a PC to which an LRL-005 wavelength meter was also connected.

Samples were pumped in lasing experiments by a 12-W IPG laser diode array (LDA) with a fibre pigtail (the fibre diameter was 100 μm). The LDA radiation was focused into a sample by a short-focus lens ($f = 5$ mm) providing the beam-waist diameter ~200 μm. The laser diode array emitted 2-ms pulses with a pulse repetition rate of 5 Hz. The emission wavelength of the LDA at room temperature was 967 nm, and it was tuned to the absorption maxima of ytterbium ions in different matrices by varying the LDA temperature. The output lasing power was measured with an EPM-1000 power meter.

Figure 3 presents the absorption spectra of the $0.7CaF_2:0.26SrF_2:0.04YbF_3$ solid-solution single crystal and of the $0.96SrF_2:0.04YbF_3$ and $0.96CaF_2:0.04YbF_3$ crystals. One can see that the shape of the absorption spectrum depends on the crystal composition (the absorption spectrum of the solid solution exhibits additional lines). The intensity of individual lines change and the zero-phonon absorption line (the 0 – 0 transition) shifts to the blue after the addition of SrF_2 to the crystal. Note that similar spectral shifts of absorption lines were also observed for neodymium ions in similar ternary fluoride systems. This allows one to obtain the required spectral properties of the rare-earth ion i.e. to control the position and intensity of absorption and luminescence lines by varying the composition of solid solutions.

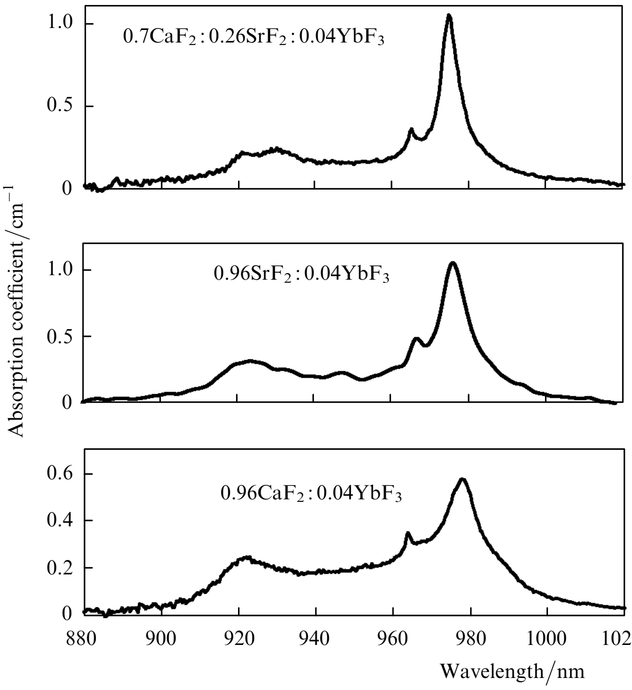

Figure 3. Absorption spectra of the $0.7CaF_2:0.26SrF_2:0.04YbF_3$, $0.96SrF_2:0.04YbF_3$ and $0.96CaF_2:0.04YbF_3$ single crystals.

Figure 4 shows the luminescence spectra of the $0.96CaF_2:0.04YbF_3$, $096SrF_2:0.04YbF_3$ single crystals and $0.7CaF_2:0.26SrF_2:0.04YbF_3$ solid solution. One can see that on passing from calcium fluoride to strontium fluoride and then to the $0.7CaF_2:0.26SrF_2:0.04YbF_3$ solid solution, the maximum of the luminescence spectrum shifts to the blue and the most intense luminescence line corresponding to the 0 – 0 transition narrows down. This is probably explained by a decrease in the number of different optical YbF_3 centres resulting in the decrease in the inhomogeneous broadening of absorption and luminescence lines. Note that a similar narrowing of the 0 – 0 line was also observed in the absorption spectrum (Fig. 3).

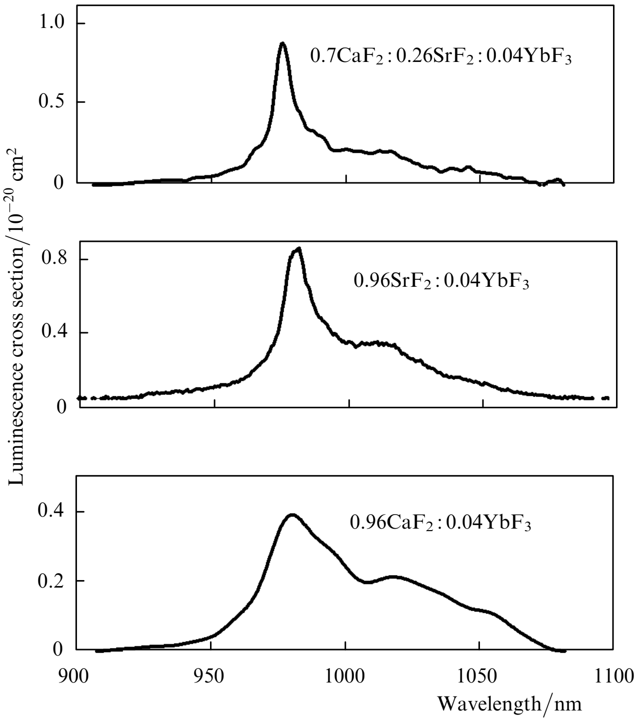

Figure 4. Luminescence spectra of the $0.7CaF_2$: $0.26SrF_2:0.04YbF_3$, $0.96SrF_2:0.04YbF_3$ and $0.96CaF_2:0.04YbF_3$ single crystals.

We measured the luminescence decay kinetics for all samples excited by nanosecond pulses from a tunable $F^{2+}:LiF$ colour centre laser. The decay kinetics was single-exponential for all single crystals studied. It was found from the decay kinetics that the lifetime of ytterbium ions at a molar concentration of 4 % was 1.8, 2.5, and 2.0 ms in CaF_2 and SrF_2 crystals, and the $0.7CaF_2:0.26SrF_2$ solid solution, respectively.

The lasing properties of ytterbium ions in fluoride single crystals were investigated in a cavity formed by a plane dichroic mirror with transmission 95 % in the wavelength range from 969 to 981 nm (the LDA emission line was

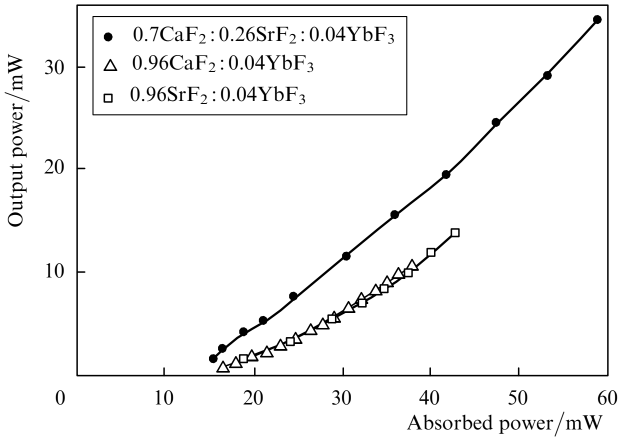

Figure 5. Dependences of the average output power of the $0.7CaF_2:0.26SrF_2:0.04YbF_3$, $0.96SrF_2:0.04YbF_3$ and $0.96CaF_2:0.04YbF_3$ single crystals on the absorbed pump power.

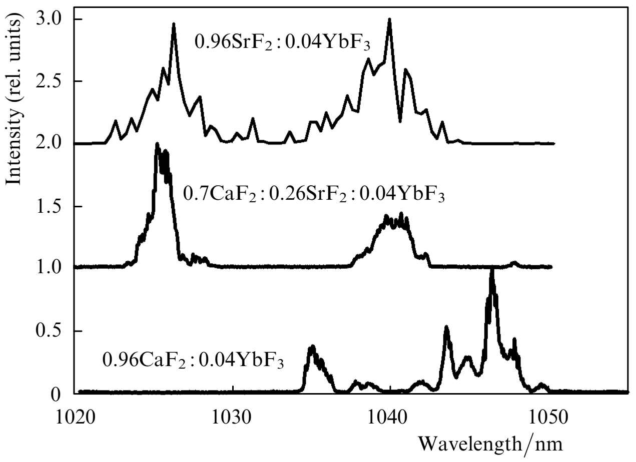

Figure 6. Lasing spectra of the $0.96SrF_2:0.04YbF_3$, $0.7CaF_2:0.26SrF_2:0.04YbF_3$ and $0.96CaF_2:0.04YbF_3$ single crystals recorded with an LRL-005 wavelength meter.

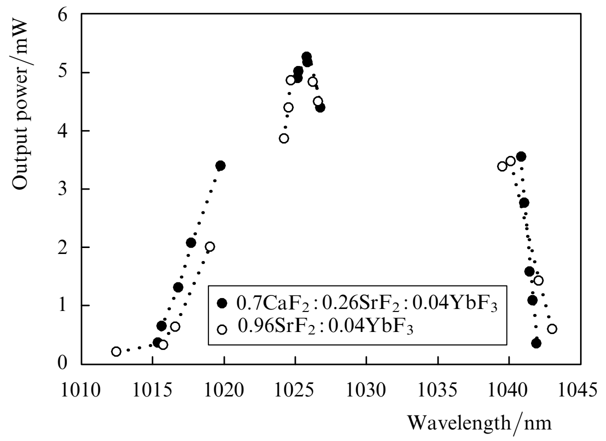

Figure 7. Output tuning curves for the $0.7CaF_2:0.26SrF_2:0.04YbF_3$ and $0.96SrF_2:0.04YbF_3$ single crystal lasers with a resonator with a selecting prism.

temperature tuned to the absorption maximum of each single crystal) and the reflectance of 100 % in the 1015–1070-nm region (Fig. 5). The output spherical mirror with the radius of curvature 100 mm was located at a distance of 45 mm from a plane dichroic mirror, i.e. the resonator configuration was close to confocal. The reflectance of the output mirror was varied from 75 % to 95 %.

Figure 5 presents the dependences of the output power of lasers based on the $0.96CaF_2:0.04YbF_3$ and $0.96SrF_2:0.04YbF_3$ single crystals and the $0.7CaF_2:0.26SrF_2:0.04YbF_3$ solid solution on the absorbed pump power for the optimal reflectance of the output mirror equal to 95 %. One can see that the solid solution has a lower lasing threshold compared to both single crystals, which is explained by its higher optical quality. The maximum efficiency for the solid-solution single crystal (with respect to the absorbed pump power) equal to 59 % was obtained for the slope efficiency equal to 83 %. The lasing threshold for the SrF_2 and CaF_2 crystals was approximately 1.5 times higher and the maximum lasing efficiency was 32 %. Note that, despite a higher lasing threshold and a lower maximum lasing efficiency of these crystals, their slope efficiency was close to that for the solid solution and amounted to 78 %.

Figure 6 shows the lasing spectra of the $0.96CaF_2:0.04YbF_3$ and $0.96SrF_2:0.04YbF_3$ single crystals and the $0.7CaF_2:0.26SrF_2:0.04YbF_3$ solid solution. One can see that the lasing spectrum of the CaF_2 crystal in the nonselective resonator is quite broad and has a maximum at ~1045 nm. For a fully aligned laser cavity, oscillations for both $Yb^{3+}:CaF_2$ crystal and $Yb^{3+}:CaF_2-SrF_2$ solid solution occurred at 1040 nm. However, the introduction of small additional losses (for example, the misalignment of the active element from the perpendicular position) leads to lasing at ~1025 nm. In this case, depending on the value of additional losses, lasing can occur simultaneously at both lines or at any of these lines.

Figure 7 presents the output tuning curves for the $0.96SrF_2:0.04YbF_3$ and $0.7CaF_2:0.26SrF_2:0.04YbF_3$ lasers with a selective resonator containing a 60° glass prism as a selective element. Both lasers can be tuned within individual lines, and lasing at the wavelength shorter than 1025 nm can occur, which can be obtained in a nonselective resonator as well. We have failed to obtained tuning between these two lines: first lasing appeared simultaneously at the two lines and then jumped to another wavelength. The shortest-wavelength lasing was obtained at 1013 nm and the longest-wavelength – at 1044 nm.

Thus, we have obtained for the first time lasing in the $0.96SrF_2:0.04YbF_3$ single crystal and the $0.7CaF_2:0.26SrF_2:0.04YbF_3$ solid solution. It is important to note that the synthesis of $CaF_2-SrF_2:YbF_3$ solid-solution single crystals is simpler than that of MeF_2: YbF_3 (Me = Ca, Sr) single crystals, which provides a higher quality of the crystal, a lower lasing threshold, and a higher lasing efficiency (the total efficiency is 53 % and the slope efficiency is 83 %). The lasing spectrum of the solid solution is close to that of the $SrF_2:YbF_3$ crystal, and short-wavelength lasing can be obtained at 1025 nm in a nonselective cavity. This allows the development of lasers with a low thermal release and a small Stokes shift between the 980-nm pump wavelength and the 1025-nm laser wavelength.

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