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Efficient laser performance of $Nd^{3+}:Sr_5(PO_4)_3F$ at 1.059 and 1.328 μm

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High efficiency, low threshold laser performance of Nd^{3+} doped $Sr_5(PO_4)_3F$, or S-FAP, crystals has been demonstrated. Its performance and properties compared with commercially available Nd:YVO_4 indicate that Nd:S-FAP is an outstanding medium for diode pumped laser applications at both 1.059 and 1.328 μm.

Low threshold, high efficiency lasing at 1.059 and 1.328 μm is reported for strontium fluorapatite, $Sr_5(PO_4)_3F$, or S-FAP, doped with Nd^{3+}. Although S-FAP crystals were successfully grown[1] more than twenty years ago, they have not been considered as useful hosts because of inadequate crystal quality. S-FAP is an apatite crystal[1] and interest in apatite crystals has been renewed recently[2–5] in the search for more efficient materials for diode laser pumping. The apatite crystals are attractive for this purpose because they are available as large, high quality crystals and in them Nd^{3+} ions have unique spectroscopic properties appropriate for diode pumping. High quality S-FAP crystals were grown by the Czochralski technique. The growing conditions are similar to those used to grow its isomorph, calcium fluorapatite (FAP) and have been discussed in detail elsewhere.[3,6]

Room temperature polarized absorption spectra of Nd^{3+}:S-FAP in the 800-nm region are given in Fig. 1. The absorption is partly polarized with the π polarization stronger than the σ polarization. The main absorption peak is centered at 805.4 nm and has a full width at half maximum of about 1.5 nm. The peak absorption coefficient is 2.74 cm^{-1} for a sample containing 0.2 mole % Nd^{3+} in the melt. Since the distribution coefficient of Nd^{3+} ions in S-FAP is 0.36,[1] the actual Nd^{3+} concentration in this crystal is estimated to be about 0.072 at. % (In what follows the Nd^{3+} concentration referred to is the actual concentration in crystal). As a result, Nd^{3+}:S-FAP has an absorption cross section of 2.26×10^{-19} cm^2. Room temperature polarized emission spectra are given in Fig. 2. This figure shows that the emission is partially polarized with the π polarization stronger than the σ polarization and that the emission is characterized by two strong lines centered at 1.059 and 1.328 μm. This unique property is believed to result from the large crystal field splitting, 368 cm^{-1}, of the $^4F_{3/2}$ manifold of Nd^{3+} ions in S-FAP. The strongest line originates from the lower sublevel of the $^4F_{3/2}$ manifold which accommodates about 86% of the total excited population at room temperature. As a result, the effective stimulated emission cross section is high as will be shown later. The room temperature emission decay of 0.072 at. % Nd^{3+}:S-FAP is exponential with a decay time of 298 μs. An S-FAP sample containing 0.68 at. % Nd^{3+} (11.4×10^{19} Nd^{3+} ions/cm^3) was used for laser testing. It has a peak absorption coefficient of 25.7 cm^{-1}. The room temperature emission decay from the $^4F_{3/2}$ manifold is nonexponential in this sample. The room temperature decay time, obtained by normalizing the fluorescence intensity at $t=0$ to one and then integrating the entire decay curve over time, was found to be only 190 μs, indicating the existence of concentration quenching in this crystal. The decay time measurements were done with fine-ground powders of crystals to avoid the decay time lengthening effect of self-absorption.

A 1.9-mm-long sample of the 0.68 at. % Nd^{3+}:S-FAP crystal was cut with flat and parallel faces containing the c axis for laser experiments. Both pulsed and cw laser-pump-laser experiments were performed in manners which simulated diode laser pumping. Pulsed laser action was excited with a long pulse Cr:$LiSrAlF_6$ (Cr:LiSAF) laser. The Cr:LiSAF laser was tuned to the Nd:S-FAP absorption peak at 805.4 nm. Its spectral bandwidth was ~1 nm. cw excitation was achieved with a cw Ti:sapphire laser tuned to 805.4 nm with spectral bandwidth of ~0.1 nm. More than 95% of the pump power was absorbed in both pump schemes. The laser resonator was composed of a 5 cm radius of curvature high reflectance (HR) mirror coated for the wavelength of interest and a flat output coupler (OC) with transmission up to 5%. The pump light was focused with a 10-cm focal length lens through the HR mirror to a point near the side of the laser crystal facing the OC.

Lasing from Nd^{3+}:S-FAP is linearly polarized along c axis and occurs at 1.0594 μm for the $^4F_{3/2}\rightarrow{}^4I_{11/2}$ transition.

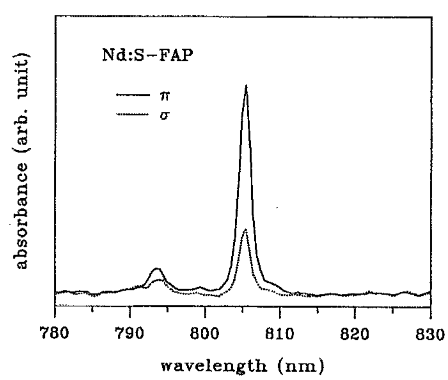

FIG. 1. Room temperature polarized absorption spectra of Nd^{3+}:S-FAP in the 800-nm region.

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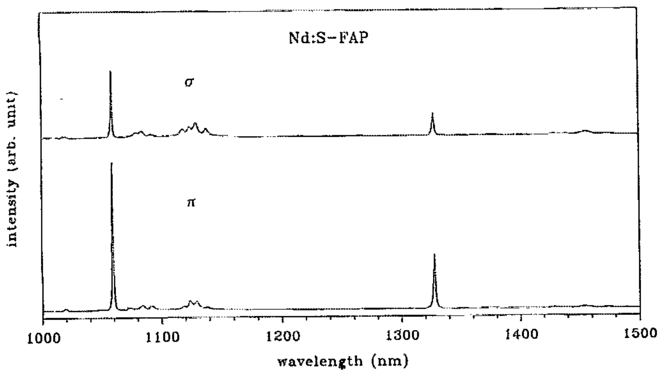

FIG. 2. Room temperature polarized emission spectra of Nd^{3+}:S-FAP in the 1.06- and 1.33-μm regions obtained by cw excitation at 805.4 nm.

The thresholds and slope efficiencies for both types of operation at this wavelength are listed in Table I for different OC transmissions. The laser output energy (power) is plotted as a function of absorbed energy (power) in Fig. 3 for a 5% OC. The measured slope efficiency can be expressed as a function of the transmission of the OC, T, as follows:

$$\eta=\eta_0 T/(T+L), \tag{1}$$

where η_0 is the intrinsic slope efficiency and L the double-pass passive loss. To best describe the results in Table I the parameters $\eta_0=71\%$ and $L=0.93\%$ can be used for pulsed operation, and $\eta_0=64\%$ and $L=0.62\%$ for cw operation.

In order to fully assess the performance of Nd:S-FAP comparison was made, using the same laser cavity, with a 2.7-mm-long Nd:YVO_4 crystal purchased from ITI Electro-Optics Corp. The emission decay time of Nd^{3+} ions in this crystal was measured to be 98 μs, which is about the same as its radiative lifetime,[7] indicating negligible concentration quenching. It has a peak absorption coefficient of 17.4 cm^{-1} at 808.7 nm. The Nd^{3+} number density was estimated to be 7×10^{19} ions/cm^3 according to the absorption data. The laser output was π-polarized and occurred at 1.0646 μm. However, the laser output power varied from region to region in the crystal, indicating inhomogeneities. The results obtained from the best performing region are summarized in Table I for comparison. The Nd:YVO_4 laser input/output characteristics are also given in Fig. 3 for a 5% OC. Comparing the lasing results, one realizes that, while the cw performances for both crystals are almost identical, the pulsed slope efficiencies for YVO_4 are slightly lower than those for S-FAP. Analysis of the slope efficiency as a function of the OC transmission for Nd:YVO_4 yields $\eta_0=68\%$ and $L=1.18\%$ for pulsed operation, and $\eta_0=64\%$ and $L=0.67\%$ for cw operation.

The absorbed power at lasing threshold can be expressed as follows:[8]

$$P_{\text{th}}=\frac{\pi h\nu_p\delta}{4\sigma_e\tau\eta_p}(w_0^2+w_p^2), \tag{2}$$

where $h\nu_p$ is the pump photon energy, η_p the pump quantum efficiency, τ the upper manifold decay time, σ_e the effective stimulated emission cross section, δ the round-trip loss which includes both internal and external losses, and w_0 and w_p the beam radii of the laser cavity mode and the pump beam, respectively. The pump quantum efficiency, the number of ions in the upper manifold created by one absorbed photon, should be near unity and therefore about the same for both crystals. In addition, w_0 and w_p are the same for both lasers since the cavity was the same for both lasers. As a result, the effective stimulated emission cross section of Nd:S-FAP can be estimated from that of Nd:YVO_4 by comparing the thresholds, the losses and the upper manifold decay times of both crystals. Taking into account the Boltzmann factor, 0.52, for the lower sublevel of the $^4F_{3/2}$ manifold in Nd:YVO_4 its effective stimulated emission cross section is found to be 10.5×10^{-19} cm^2 (Ref. 9). We therefore estimate the effective stimulated emission cross section of Nd:S-FAP to be 5.4×10^{-19} cm^2.

Figure 2 shows that the relative branching ratio of the $^4F_{3/2}\rightarrow{}^4I_{13/2}$ transition at around 1.3 μm to the $^4F_{3/2}\rightarrow{}^4I_{11/2}$ transition at around 1.06 μm is quite high. The ratio of the peak intensities at 1.3279 and 1.0594 μm was measured to be 0.37. The similar ratio is only 0.24 in Nd:YVO_4, which has been considered to be one of the best 1.3 μm lasers.[9] We tested 1.3-μm lasing using the same setup described above except for the coatings. Both mirrors of the resonator in this case have high transmission (~90%) at around 1.06 μm. The crystal was antireflection coated at both 1.06 and 1.33 μm. Lasing output was π polarized and occurred at 1.3279 μm. The input/output lasing characteristics obtained with a 5% output coupler are given in Fig. 3(a) for pulsed and in 3(b) for cw operation. The cw threshold and slope efficiency are 14 mW and 46%, respectively. In pulsed operation the threshold and slope efficiency are 22 μJ and 52%. The quantum limited slope efficiency is about 60% for the 1.33-μm transition, and so it is clear that Nd:S-FAP performs very well as a 1.33-μm laser. In fact, to our knowledge, this is the best performance ever reported for the Nd^{3+} 1.3-μm lasers.

TABLE I. Comparison of laser performance for Nd:S-FAP at 1.0594 μm and Nd:YVO_4 at 1.0646 μm in pulsed and cw operation.

	Threshold				Slope efficiency (%)			
	Pulsed (μJ)		cw (mW)		Pulsed		cw	
T (%)	S-FAP	YVO_4	S-FAP	YVO_4	S-FAP	YVO_4	S-FAP	YVO_4
1.2	5	3	4	4	40	34	42	41
3	6	4	5.5	4.6	54	49	54	53
5	9	6	6	6	60	54	59	59

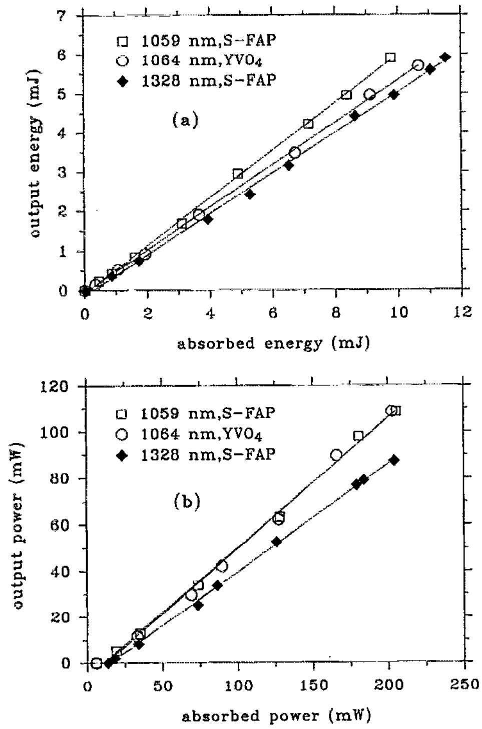

FIG. 3. The input/output characteristics of Nd^{3+}:S-FAP and $Nd^{3+}:YVO_4$ for (a) pulsed and (b) cw operation.

By comparing the thresholds at 1.059 and 1.328 μm the effective stimulated emission cross section at 1.328 μm was estimated to be not less than 2.3×10^{-19} cm^2.

The recent interest in Nd:YVO_4 results from the facts that it has a high absorption cross section and a large product of the effective emission cross section (σ_e) and the decay time (τ_r). A high absorption cross section makes it easy to obtain single-mode operation by using a short monolithic cavity.[10] The absorption cross section data reported in the literature for Nd:YVO_4 are not consistent. According to the absorption coefficient given in Ref. 10 for two samples with different concentrations the absorption cross section ranges from 2.10 to 2.87×10^{-19} cm^2, while Ref. 11 implies a value of only 2.0×10^{-19} cm^2. Therefore the absorption cross section of Nd:S-FAP, 2.26×10^{-19} cm^2, is at least comparable to, if not higher than, that of Nd:YVO_4.

Since the lasing threshold power is inversely proportional to $\sigma_e\tau$ as seen in Eq. (2), a large $\sigma_e\tau$ assures low threshold operation suitable for diode pumping. When concentration quenching does not exist in the crystal τ reaches its maximum value τ_r, the radiative lifetime. As a result, the $\sigma_e\tau$ product has its highest value, $\sigma_e\tau_r$. The estimated $\sigma_e\tau_r$ product for Nd:S-FAP is 1.6×10^{-19} cm^2 ms and is about 1.5 times as large as that of Nd:YVO_4, 1.1×10^{-19} cm^2 ms. As mentioned above, concentration quenching exists in the S-FAP crystal used in our lasing experiment but it is negligible in the tested YVO_4 sample. However, both crystals performed almost identically as 1.06-μm lasers in our experiments. It is therefore reasonable to expect that an Nd^{3+} concentration for S-FAP can be found where $\sigma_e\tau$ exceeds that of YVO_4 and which results in a material with superior diode pumped laser performance.

It has been very difficult to grow large, high quality Nd:YVO_4 crystals.[9] As mentioned earlier we observed that the laser performance varied from point to point in the crystal, changing the output power by as much as 50% between adjacent sites. This indicates inhomogeneous crystal quality. In addition, the Nd:YVO_4 crystal was very easily damaged in long-pulsed operation. Pump-laser-induced bulk crystal damage in Nd:YVO_4 developed in time and usually occurred at lower pump fluences at sites which did not lase well. Such inhomogeneities and damage phenomena were not observed in S-FAP, which is much easier to grow into high quality, large crystals. However, in S-FAP inclusions sometimes appear in the core region of the crystal boule. Research to eliminate this problem is in progress.

In summary, we have demonstrated the excellent lasing potential of the Nd^{3+}-doped strontium fluorapatite crystal, Nd:S-FAP, at both 1.059 and 1.328 μm. The large absorption cross section, high emission cross section, long radiative lifetime, and most importantly, the availability of high quality, low cost crystals should make Nd:S-FAP a very attractive material for diode-laser-pumped laser applications. This is particularly so for Nd:S-FAP as a 1.3-μm laser.

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