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New Ytterbium-doped Apatite Crystals for Flexible Laser Design

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ABSTRACT

A new class of Yb-lasers is summarized in this article. The apatite family of crystals has been found to impose favorable spectroscopic and laser properties on the Yb^{3+} activator ion. Crystals of Yb-doped $\text{Ca}_5(\text{PO}_4)_3\text{F}$, $\text{Sr}_5(\text{PO}_4)_3\text{F}$, $\text{Ca}_x\text{Sr}_{5-x}(\text{PO}_4)_3\text{F}$, and $\text{Sr}_5(\text{VO}_4)_3\text{F}$ have been grown and investigated. Several useful laser crystals have been identified which offer a variety of fundamental laser parameters for designing diode-pumped systems. In general, this class of materials is characterized by high emission cross sections ($3.6 - 13.1 \times 10^{-20} \text{ cm}^2$), useful emission lifetimes (0.59 - 1.26 msec), a strong pump band ($\sigma_{\text{abs}} = 2.0 - 10.0 \times 10^{-20} \text{ cm}^2$) and pump and extraction wavelengths near 900 and 1045 nm, respectively. Efficient lasing has been demonstrated for several of the members of this class of materials, and high optical quality crystals have been grown by the Czochralski method.

1. INTRODUCTION

Yb^{3+} has long been recognized to serve as a laser ion when incorporated as a dopant into $\text{Y}_3\text{Al}_5\text{O}_{12}$ and other garnets.¹ The main obstacle to further development entailed the limited pump absorption features that are available with Yb-doped crystals. Since Yb^{3+} is a $4f^{13}$ ion, it possesses only two relevant electronic states — the $^2\text{F}_{7/2}$ ground state and the $^2\text{F}_{5/2}$ excited state — separated by about $10,000 \text{ cm}^{-1}$. Since the prevailing pump sources in the 1960s and 1970s only included broad white light generating flashlamps, the single narrow absorption feature of Yb^{3+} proved to be an inadequate means of effectively pumping the laser material. While the need for a useful narrow band pump source was recognized some time ago,² it was not until the emergence of InGaAs laser diodes that some workers recognized the renewed significance of Yb-doped materials.³ In fact, with the availability of commercial laser diodes, the limited absorption

features of Yb actually provide an advantage, since the lack of higher-lying excited states assures the absence of detrimental processes such as upconversion and excited state absorption. Additionally, the Yb^{3+} dopant offers an important advantage over the most common laser ion, Nd^{3+} , since its emission lifetime tends to be about four times greater when the same host medium is considered for both ions, thereby allowing for an enhanced level of energy storage.

The main complication associated with solid-state lasers based on the Yb ion involves the quasi-four-level nature of the system at room temperature. Otherwise stated, the problem is that the terminal laser level tends to reside at an energy of $200\text{--}600 \text{ cm}^{-1}$, which is comparable to the thermal energy present at room temperature (200 cm^{-1}).⁴ Accordingly, the search for new Yb^{3+} lasers has been intimately tied to the structure of the energy levels and the strength of the transitions among them. In particular, the nature of the crystal field splitting in the $^2\text{F}_{7/2}$ and $^2\text{F}_{5/2}$ states and the cross section associated with the various $^2\text{F}_{7/2} - ^2\text{F}_{5/2}$ transitions turns out to have a crucial impact on the viability of the system as a laser material. In brief, the ideal Yb-laser would possess a high-lying crystal field component in the $^2\text{F}_{7/2}$ ground state, to which a particularly intense transition from the $^2\text{F}_{5/2}$ excited state would exist. The Yb-doped apatite class of laser materials to be described in this article is characterized by just such an energy level structure along with the desired transition strength moments.⁵

As a result of our recent efforts, the Yb^{3+} -doped apatite-structure crystals may now potentially be employed in a variety of diode-pumped laser systems. This family of laser materials offers a substantial range of fundamental laser parameters with which systems may be designed, in order to adjust the pumping, storage, and energy extraction characteristics. In other words, the Yb-doped

apatite class of materials allows a number of trade-offs, wherein the emission lifetime and cross section, as well as the pump line width and saturation parameters can be altered, albeit not independently of each other. A range of possibilities now exists such that the specific parameters of a given Yb:apatite laser medium can be matched to the requirements of a particular application. Among the crystals that have turned out to be useful Yb laser materials are $\text{Ca}_5(\text{PO}_4)_3\text{F}$ (FAP),⁵ $\text{Sr}_5(\text{PO}_4)_3\text{F}$ (S-FAP),⁶ $\text{Ca}_3\text{Sr}_2(\text{PO}_4)_3\text{F}$ (C_3S_2 -FAP),⁷ and $\text{Sr}_5(\text{VO}_4)_3\text{F}$ (S-VAP).⁸ It is important to note that the apatite crystals were originally investigated as Nd laser host media;⁹ our contribution was to recognize that the Yb ion exhibits particularly useful laser properties in these crystals. It is also noteworthy that Yb:YAG has also proved to be a highly effective diode-pumped laser material, as discussed recently.¹⁰

2. SPECTROSCOPY

The measured absorption and emission cross sections, σ_{abs} and σ_{em} , and the upper level lifetimes, τ_{em} , can be used to assess several important laser performance parameters for different Yb:apatite lasers. In particular, β_{min} , which is defined as the minimum fraction of Yb ions that must be excited in order to balance the gain and the ground state absorption at the extraction wavelength, is crucial in quasi-four-level systems such as Yb^{3+} . The β_{min} parameter may be determined then from the absorption and emission cross section values for the extraction wavelength of the laser, λ_{ext} , according to:³⁻⁵

$$\beta_{\text{min}} = \frac{\sigma_{\text{abs}}(\lambda_{\text{ext}})}{\sigma_{\text{abs}}(\lambda_{\text{ext}}) + \sigma_{\text{em}}(\lambda_{\text{ext}})} . \quad (1)$$

The β_{min} parameter may also be expressed in terms of the individual transitions between the crystal field components of the electronic states.³⁻⁵

Another performance parameter which can be assessed from the spectroscopic properties is the pump saturation intensity, I_{sat} . For efficient diode pumping, a large fraction of Yb ions must be excited in order to overcome the ground state absorption losses, and I_{sat} is a measure of the ease of bleaching the Yb-doped crystal. This pump parameter is assessed from the σ_{abs} value at the pump wavelength, λ_{p} , and the emission lifetime, τ_{em} , using:

$$I_{\text{sat}} = \frac{hc}{\lambda_{\text{p}} \sigma_{\text{abs}} \tau_{\text{em}}} . \quad (2)$$

Clearly, higher absorption at the pump wavelength and longer Yb lifetimes are desirable for low values of I_{sat} .

Finally, an important parameter which takes into account both the absorption and emission characteristics of the Yb-laser crystal is I_{min} . In the case of lightly-doped crystals, I_{min} is the minimum absorbed pump intensity which is required to reach threshold (in an otherwise lossless oscillator). This parameter is simply the product of our two previous laser performance indicators or:

$$I_{\text{min}} = \beta_{\text{min}} I_{\text{sat}} . \quad (3)$$

Lower values of I_{min} imply lower threshold lasers and therefore are more desirable when all else considered is equal.

Figure 1. Absorption and emission spectra of Yb in $\text{Sr}_5(\text{VO}_4)_3\text{F}$ (S-VAP), with E||c, on absolute cross section scale.

Figure 2. Absorption and emission spectra of Yb:Sr₅(VO₄)₃F with E ⊥ c.

The polarized absorption and emission cross sections of Yb:S-VAP are plotted for E||c and E ⊥ c in Figs. 1 and 2, respectively. Interestingly, the uniaxial nature of the apatite-structure has led to highly anisotropic, favorable spectral features. In particular, for light polarized along the c-axis of the crystal, the absorption spectrum is dominated by a single, strong feature at 905 nm. Similarly, in the emission spectrum of E||c, the oscillator strength is largely concentrated into the feature at 1044 nm. The high values of σ_{abs} and σ_{em} observed for these two peaks suggest useful pump and extraction parameters for an Yb:S-VAP laser. We have previously observed similar useful attributes in crystals of Yb:FAP⁵ and Yb:S-FAP.⁶ More recently, spectral determinations have additionally been completed⁷ for several mixed crystals of Ca_{5-x}Sr_x(PO₄)₃F:Yb where $x = 1, 2$ and 3 (referred to as CS-FAP). As a useful way to compare the potential laser performance of known Yb-doped apatite crystals, we display data points on a figure-of-merit plot with σ_{em}

on the ordinate and I_{min} on the abscissa. Such a plot is presented in Fig. 3 and includes the data points of several Yb-doped crystals which we have presented in an earlier article.⁴ This plot very clearly demonstrates that the favorable high σ_{em} , low I_{min} space is uniquely occupied by the various apatite structure crystals we have been investigating.

Figure 3. Plot of the extraction cross section (σ_{ext}) against the minimum absorbed pump intensity required to yield net gain (I_{min}). All of the Yb-doped apatite-structure crystals are located in the superior upper-left-hand sector of this plot.

3. LASER PERFORMANCE

The cw laser performance was assessed for all but one of the crystals that were spectroscopically characterized in the last section. Here we present data to prove that the Ca-Sr mixed apatite crystals, as well as the new fluorovanadate host, serve as useful laser media. Furthermore, the prior results obtained for Yb:FAP⁵ and Yb:S-FAP⁶ will be briefly reviewed. Overall, four new Yb-based laser crystals have been recognized to lase effectively.

Since we have already described the techniques employed to measure the laser efficiencies,^{5,6} only the highlights of our methods will be outlined here. The Yb-doped crystals were cut to 0.5-1.0 cm in length with parallel uncoated windows, and were arranged to be at the center of a 10 cm long concentric cavity. The Ti:sapphire pump laser output was

set to 900-905 nm, and chopped to a duty cycle of 25% before impinging on the crystal (although operating in the true cw mode appeared to have little or no impact on the Yb-laser output). Using this experimental approach, we have produced laser action in Yb:S-VAP ($\text{Sr}_5(\text{VO}_4)_3\text{F}$), Yb:C₄S-FAP ($\text{Ca}_4\text{Sr}(\text{PO}_4)_3\text{F}$), and Yb:C₃S₂-FAP ($\text{Ca}_3\text{Sr}_2(\text{PO}_4)_3\text{F}$). Several examples of the laser data obtained for Yb:S-VAP are displayed in Fig. 4, where the output couplings employed ranged from 4.4% to 19.8%, yielding slope efficiencies of 36.6, 56.4, and 62.9%, with respect to the absorbed pump power. Several additional plots of the output power versus absorbed pump power appear in Fig. 5, including Yb in both of the mixed apatite hosts, C₃S₂-FAP and C₄S-FAP, operating near the standard 1046 nm wavelength.

Figure 4. Laser efficiency data acquired for Yb:S-VAP for the three indicated output coupler values at 1044 nm with E||c.

Figure 5. Illustrative laser efficiency results obtained for the mixed apatites, including Yb:C₃S₂-FAP at 1046 nm, and Yb:C₄S-FAP at 1045, 1110, and 985 nm (output coupling values are denoted on the figure).

Interestingly, we were able to also lase the Yb:C₄S-FAP crystal at 1110 nm, which corresponds to a longer wavelength, weaker emission band. We expect that all of the Yb-doped apatite crystals are amenable to laser action near this longer wavelength. It is noteworthy that, in spite of the low cross section value, the generation of 1110 nm output is somewhat enhanced by the fact that true four-level operation is possible at this wavelength (i.e., no ground state absorption is present). Note that Yb:C₄S-FAP has also been demonstrated to lase at 985 nm (with E_Lc). Since this spectral line corresponds to the transition between the lowest crystal field components of the $^2\text{F}_{7/2}$ ground and $^2\text{F}_{5/2}$ excited states, these experiments correspond to true three-level operation of this system. Therefore the 985, 1046 and 1110 nm operating wavelengths of Yb:C₄S-FAP correspond to three, quasi-four, and four-level operation of the laser!

The laser data can be analyzed further on the basis of the simple equation describing the slope efficiency, η , in the limit of low (double-pass) loss, L_d , and low output coupling, T :

$$\eta = \eta_0 \frac{T}{T + L} \quad (4)$$

Here, η_0 provides a measure of the so-called intrinsic efficiency and is, in principle, simply given by the ratio of the pump and laser wavelengths (about 86% for the Yb:apatite crystal family). A summary of the single-pass losses and intrinsic efficiencies obtained for all of the Yb:apatite laser crystals is summarized in Table I, where the η_0 values of 65-84% are all in accord with the imposed fundamental quantum limit of 86% — the tendency to not attain the theoretical maximum is most likely a consequence of inadequate mode-matching of the pump and cavity fields. The range of single-pass losses from 0.7 to 3.0% is the result of variation in the quality of optical materials that have been employed. On the basis of the data presented in this section, the laser performance of the Yb-doped apatite crystals is judged to be good at this time, and may be expected to improve further as lower loss material becomes available.

Table I. Summary of laser efficiency analysis for Yb-doped apatite crystals

Crystal	Wavelength (nm)	Intrinsic efficiency (%)	Single-pass loss (%)	Path length (cm)
S-VAP	1044	83	3.0	0.57
FAP ^a	1043	84	1.2	0.69
C ₄ S-FAP	1046	76	1.7	0.75
	1110	81	2.7	
C ₃ S ₂ -FAP	1046	65	1.8	0.65
S-FAP ^b	1047	70	0.7	0.70

^aRef. 6

^bRef. 7

4. CONCLUSION

We have defined a new class of laser media based on the Yb³⁺ ion that offers a substantial range of laser parameters. The emission cross section was found to be as low as $3.6 \times 10^{-20} \text{ cm}^2$ for Yb in the Ca₄Sr(PO₄)₃F (C₄S-FAP) host, and as large as $13.1 \times 10^{-20} \text{ cm}^2$ for Yb:Sr₅(VO₄)₃F (S-VAP). The emission cross section variations are, of course, directly tied to changes in the emission lifetime (0.59-1.26 msec). All of the crystals have been demonstrated to lase efficiently and can be grown as high-quality optical materials by the Czochralski method. We believe that the Yb:apatite class of lasers is useful for low and medium-power applications, and will be preferred over the more common Nd-lasers in many situations by virtue of their longer lifetime.

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