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## New criteria to choose the best Yb<sup>3+</sup>-doped laser crystals

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PACS. 42.55.Ah – General laser theory. PACS. 42.55.Xi – Diode-pumped lasers.

**Abstract.** – Using a model for the quasi-three-level laser dealing with Gaussian waves, taking into account the saturation of the pump, the stimulated emission at the pump wavelength, the variation of the pump and laser waists along propagation (important for laser diode pumping) and the variation of the laser intensity along propagation, we evaluate the more promising Yb<sup>3+</sup>-doped crystals for laser applications: YAG, LNB, GdCOB, YCOB, YAB, C-FAP, S-FAP, CLYPA, KGdW, KYW, Sc<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, Lu<sub>2</sub>O<sub>3</sub>. Our evaluation, established for the CW oscillator regime, differs from that of Deloach and coworkers valid for ns pulse extraction mode. KYW, KGdW and Sc<sub>2</sub>O<sub>3</sub> are found to be the most efficient when considering the laser extracted power and S-FAP, C-FAP, YAG, KYW and KGdW are the most efficient when considering the small-signal gain of the amplifier regime.

Introduction. – The most promising ion that can be used in a non-Nd laser in the same range of emission wavelength is Yb<sup>3+</sup>. The Yb<sup>3+</sup> ion have some advantages over the Nd<sup>3+</sup> ion as laser emitting center due to its very simple energy level scheme, constituted of only two levels: the  ${}^{2}F_{7/2}$  ground state and the  ${}^{2}F_{5/2}$  excited state. There is no excited-state absorption reducing the effective laser cross-section, no up-conversion, no concentration quenching and no absorption in the green, which is very favourable in the case of a self-frequency doubling laser (SFD). The intense Yb<sup>3+</sup> absorption lines are well suited for laser diode pumping near 980 nm and the small Stokes shift (about 650 cm<sup>-1</sup>) between absorption and emission reduces the thermal loading of the material during laser operation. The disadvantage of Yb<sup>3+</sup> is that the final laser level is thermally populated (quasi-three-level laser), increasing the threshold.

In the last decade, the output power of Yb<sup>3+</sup>-doped lasers has exponentially increased, up to 434 W in a longitudinal pumping configuration at Livermore [1] and very recently up to 1.08 kW [2], 346 W at Stuttgart [3] and more than 1 kW in side-pumped configuration at Hughes [4]. Furthermore, several Yb lasing in SFD materials have been reported [5–9] and in one paper [10], SFD has been obtained (in LiNbO<sub>3</sub> : MgO : Yb (LNB), 60 mW green, the best efficiency ever reported for a non-Nd SFD laser).

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 $Ca_5(PO_4)_3F$  (C-FAP) and  $Y_3Al_5O_{12}(YAG)$  were soon recognized to be favourable hosts for Yb lasing. This fact was supported by an evaluation [11], valid for ns pulse extraction mode, of the spectroscopic properties of several Yb-doped crystals useful for laser action. This evaluation is based on two parameters known from spectroscopy: the emission crosssection  $\sigma_e$  at the laser wavelength and the minimum pump intensity  $I_{min}$  required to achieve transparency at the laser wavelength:

$$I_{\min} \frac{\sigma_{\rm a}}{\sigma_{\rm e} + \sigma_{\rm a}} \frac{h\nu}{\sigma_{\rm ap}\tau},\tag{1}$$

where  $\sigma_a$  is the absorption cross-section at the laser wavelength,  $\sigma_{ap}$  is the the absorption cross-section at the pump wavelength and  $\tau$  is the  ${}^2F_{5/2}$  lifetime.  $\sigma_e$  and  $I_{min}$  were used in [11] as figure of merit to classify the hosts, in a two-dimensional diagram similar to fig. 1, where we introduced new promising hosts (spectroscopic data in [12–19]) recently tested: Sr<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>F (S-FAP), KY(WO<sub>4</sub>)<sub>2</sub> (KYW), KGd(WO<sub>4</sub>)<sub>2</sub> (KGdW), CaGd<sub>4</sub>(BO<sub>3</sub>)<sub>3</sub>O (GdCOB), CaY<sub>4</sub>(BO<sub>3</sub>)<sub>3</sub>O (YCOB), YAl<sub>3</sub>(BO<sub>3</sub>)<sub>4</sub> (YAB), LiNbO<sub>3</sub> : MgO (LNB), Sc<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, Lu<sub>2</sub>O<sub>3</sub>. In the diagram, C-FAP and S-FAP appear to be exceptionally good, and YAB, GdCOB and YCOB appear modest. This is somewhat in contradiction with experimental laser tests in which these latter materials have been revealed very efficient: 73% slope efficiency in YAB, 77% in GdCOB, 78% in KYW, 72% in KGdW, to be compared to 71 and 79% for S-FAP and C-FAP, respectively. We think that there is a need for a new evaluation of Yb-doped crystals in order to predict the laser efficiency in CW extraction mode.

The purpose of this work is to present a new evaluation based on a quasi-three-level laser model, checked to be close to experimental laser data. The model deals with Gaussian waves, takes into account the saturation of the pump (which occurs for the Yb ion because the  ${}^{2}F_{5/2}$  level has a long lifetime up to 2.5 ms and accumulates population), the stimulated emission at the pump wavelength, the variation of the pump and laser waists along propagation (important for laser diode pumping) and the variation of the laser intensity along propagation. It is a generalisation of simpler previous models as the one of Risk [20] and it predicts more accurately the laser output power and the small-signal gain.

Model of quasi-three-levels laser. – We suppose that the laser crystal and the waves propagation extend along the z-axis from z = 0 to z = L. The radial coordinate is r. The CW end-pumping occurs at a wavelength  $\lambda_p$  near 980 nm for Yb<sup>3+</sup>. In the case of a laser oscillator, the input mirror is located at z = 0 and the concave output mirror at z = d. In the case of a laser amplifier, there is of course no mirror. The fractions of population of the laser level ( ${}^2F_{5/2}$ ) and of the ground state ( ${}^2F_{7/2}$ ), N and  $N_0$  respectively, in a steady state, are such that

$$N_{0}(r, z) + N(r, z) = 1,$$
  
$$0 = -\frac{N}{\tau} + I_{\rm p}(r, z) \big(\sigma_{\rm ap} N_{0} - \sigma_{\rm ep} N\big) + I_{\rm L}(r, z) \big(\sigma_{\rm a} N_{0} - \sigma_{\rm e} N\big),$$
(2)

where  $\tau$  is the laser level lifetime,  $I_{\rm p}(r, z)$ ,  $I_{\rm L}(r, z)$  are, respectively, the densities (photons/(cm<sup>2</sup> s)) of the pump beam and of the laser waves inside the crystal (there are two counter-propagating laser waves in the oscillator case and only one forward propagating laser wave in the amplifier case).  $\sigma_{\rm ep}$ ,  $\sigma_{\rm ap}$  are, respectively, the emission and absorption cross-sections of the pump and  $\sigma_{\rm e}$ ,  $\sigma_{\rm a}$  the emission and absorption cross-sections of the laser beam.

The equation of propagation for the  $\psi_1^+(r,z) \exp[-k_1 z]$  electric field of the laser wave

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Fig. 1 – Figure of merit from ref. [11] for several promising Yb-doped hosts.

Fig. 2 – Laser oscillator output yield  $(P_{out}/P_{pump})$  and amplifier small-signal gain predicted by the model discussed in the second section for different crystals.

stands in a nonparabolic approximation as

$$\frac{\partial \psi_1^+}{\partial z} + \frac{i}{2k_1} \nabla_t^2 \psi_1^+ = \left[ \sigma_{\rm e} N(r, z) \frac{C}{2} - \sigma_{\rm a} N_0(r, z) \frac{C}{2} - \frac{\alpha_1}{2} \right] \psi_1^+ , \qquad (3)$$

where C is the laser ion concentration. Equation (3) is valid for forward propagation; for backward propagation ( $\psi_1^-$  electric field), we have to change the signs of  $k_1$  and of the bracket. The second term of the left-hand side of (3) describes the diffraction, the first term of the right-hand side of (3) describes the laser gain (calculated with the help of (2)), the second term describes the laser absorption by the Yb<sup>3+</sup> ions and the third term represents the losses from the crystal.

The equation of propagation for the  $\psi_{\rm p}(r,z) \exp[-k_{\rm p}z]$  electric field of the pump wave stands as

$$\frac{\partial \psi_{\rm p}}{\partial z} + \frac{i}{2k_{\rm p}} \nabla_t^2 \psi_{\rm p} = \left[ \sigma_{\rm ep} N(r, z) \frac{C}{2} - \sigma_{\rm ap} N_0(r, z) \frac{C}{2} \right] \psi_{\rm p}; \tag{4}$$

the Gaussian solutions of (3), (4) are given, respectively, by

$$\psi_1^+(r,z) = \alpha_1^+(z) E_{01}^+ \exp\left[G_1^+(z) + i\phi_1^+(z) + iQ_1^+(z)r^2\right],\tag{5}$$

$$\psi_{\rm p}(r,z) = \alpha_{\rm p}(z) E_{0\rm p}^+ \exp\left[G_{\rm p}(z) + i\phi_{\rm p}(z) + iQ_{\rm p}(z)r^2\right].$$
(6)

In (5), (6) we have the complex functions  $Q_j^+ = Q_j^{'+} + iQ_j^{''+}$  (j = 1, p) whose real part is related to the radius of curvature of the beam and the imaginary part to the waist of the beam  $(Q_j'' = 1/W_j^2)$ , and we have the auxiliary functions  $\alpha$  chosen as in the familiar Gaussian optics to fulfill:

$$\frac{1}{\alpha_1^+} \frac{\mathrm{d}\alpha_1^+}{\mathrm{d}z} = \frac{2}{k_1} Q_1^+, \qquad \frac{1}{\alpha_\mathrm{p}} \frac{\mathrm{d}\alpha_\mathrm{p}}{\mathrm{d}z} = \frac{2}{k_\mathrm{p}} Q_\mathrm{p},\tag{7}$$

with the initial condition  $\alpha_i^+(0) = 1$ .

	Conc. $(10^{20} \text{ ions/cm}^3)$	$ \overset{\sigma_{\rm ap}}{(10^{-20}{\rm cm}^2)} $	$ \overset{\sigma_{\rm ep}}{(10^{-20}{\rm cm}^2)} $	$ \overset{\sigma_{\rm e}}{(10^{-20}{\rm cm}^2)} $	$\frac{\sigma_{\rm a}}{10^{-20}{\rm cm}^2}$	
YAG	8.97	0.8	0.159	2.03	0.149	
YCOB	8.97	1.0	0.959	0.36	0.026	
GdCOB	8.97	1.1	1.12	0.5	0.027	
YAB	8.97	3.4	2.99	0.8	0.040	
C-FAP	0.36	10.0	0.132	5.9	0.377	
S-FAP	0.36	8.6	0.076	7.3	0.407	
CLYPA	0.36	13.0	0.229	3.8	0.164	
KGdW	8.97	12	14.6	2.7	0.289	
KYW	8.97	13.3	16.0	3	0.299	
LNB	1.89	1	1.02	1	0.023	
$Sc_2O_3$	8.97	4	4.0	1.2	0.05	
$Y_2O_3$	8.97	2.4	2.3	0.85	0.06	
$Lu_2O_3$	8.97	3	2.9	1.07	0.07	
	$\lambda_{ m p}$	$\lambda_{ m e}$	au	$L_{\mathrm{opt}}$	$R_{\mathrm{opt}}$	$L_{\rm opt}$
	(nm)	(nm)	$(\mu \mathrm{s})$	OSC (mm)	OSC (%)	AMP (mm)
YAG	942	1029	951	2.9	93	4.5
YCOB	976.4	1032	2280	3.4	98	7.1
GdCOB	975	1035.3	2500	3.4	98.5	7.4
YAB	975	1040.3	680	1.2	99.5	3.0
C-FAP	905	1043.3	1100	8.5	95	17.6
S-FAP	899	1047.3	1260	9.5	92	19
CLYPA	902	1041.7	850	7.3	98	16.4
KGdW	981	1023.3	600	0.45	99.5	1.9
KYW	981.2	1025.3	600	0.4	99.5	1.8
LNB	980	1060	540	9	98	13.4
$\mathrm{Sc}_2\mathrm{O}_3$	975	1041	800	1.2	99.5	3.2
$Y_2O_3$	975	1031	850	1.6	99	3.6
$\mathrm{Lu}_2\mathrm{O}_3$	975	1032	820	1.4	99.5	3.4

TABLE I – Spectroscopic data of Yb-doped crystal evaluated in this work.

After inserting (5) in (3), we multiply the two sides of eq. (3) by  $\psi_{\rm p}^{+*}(r, z)dx dy$  and integrate over all the *xy*-plane at position *z*. For the pump wave, after inserting (6) in (4), we multiply the two sides of eq. (4) by  $\psi_{\rm p}^{*}(r, z)dx dy$  and integrate over all the *xy*-plane at position *z*. The result with more details can be found elsewhere [21].

Evaluation of Yb<sup>3+</sup>-doped crystals. – Our evaluation is based on the output power extracted from a laser oscillator and on the small-signal gain of a laser amplifier, both being calculated with the model of the previous section. The crystals which are considered are: YAG, YCOB, GdCOB, C-FAP, S-FAP, CLYPA (Ca<sub>4</sub>La(PO<sub>4</sub>)<sub>3</sub>O, Y stands for Yb), YAB, KGW, KYW, Sc<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, Lu<sub>2</sub>O<sub>3</sub>.

In the oscillator case, the crystals were located inside the same laser cavity. The pump power has been fixed to 1 W, the chosen cavity was the one of ref. [22] concerning YAG : Yb, with a beam waist of  $22 \,\mu\text{m}$  and a pump waist of  $29 \,\mu\text{m}$ . In the amplifier case, the pump power has been fixed to 1 W, the waists of the pump and of the small signal injected have been fixed to  $29 \,\mu\text{m}$ . The power of the injected signal was 1% of the pump.

The crystals concentration was chosen typical in laser experiments for each crystal. We used a rather high concentration:  $8.97 \cdot 10^{20}$  ions/cm<sup>3</sup> for crystals having a lanthanide crys-

Crystal	YAG	GGG	C-FAP	S-FAP	GdCOB	YAB	KGW	$\mathrm{Sc}_2\mathrm{O}_3$	$Y_2O_3$
Thermal conductivity									
$W/(m ^{\circ}C)$	13	12	2	2	2.2	3-4	3.5	15.5	12.8

TABLE II – Thermal conductivity of the most promising laser crystals.

tallographic site (YAG, YCOB, GdCOB, YAB, KGdW, KYW), and a lower concentration:  $0.36 \cdot 10^{20}$  ions/cm<sup>3</sup> [11] for crystals having a low segregation coefficient (C-FAP, S-FAP and CLYPA). Let us notice that in the CLYPA case, the Yb<sup>3+</sup> segregation coefficient is very low: 0.03 and then CLYPA bulk crystals with the concentration used in this work have not been grown at the present time [23].

The crystal spectroscopic parameters used are found in the literature (see [12–19,23]) and are gathered in table I. For each crystal we have determined numerically (table I) with the model the crystal length  $L_{\text{opt}}$  (in both the oscillator and amplifier cases) and the reflectivity  $R_{\text{opt}}$  of the output mirror (in the oscillator case) leading to the maximum laser output power.

The results of calculation are shown in fig. 2. We can see that KGdW, KYW,  $Sc_2O_3$  and YAG have a high potential. The first rank of crystals given by such evaluation depend on the application (oscillator or amplifier). Tungstate can be grown either by the top nucleated floating crystal method [24,25] or from the flux by a modified Czochraski method [14], whereas lanthanide sesquioxides can be elaborated by laser heated pedestal growth (LHPG) [26–28] and by the Bridgman method using a rhenium crucible [17].

In the evaluation of an Yb-doped laser crystal, we have also to keep in mind another factor: the thermal conductivity which plays an important role in the elimination of the thermal loading of the crystal during laser operation (the laser is a quasi-three-level one). In table II, we have gathered the thermal conductivity of the most promising laser crystals. From this point of view,  $Sc_2O_3$ ,  $Y_2O_3$ ,  $Lu_2O_3$  cubic sesquioxides which show the highest thermal conductivity have to be considered. Let us point out that the epitaxial growth of undoped KYW on an Yb-doped KYW crystal is in progress in our laboratory by G. Métrat, in order to evacuate the thermal loading inside the doped crystal during laser operation.

Conclusion. – We have presented an accurate model for a quasi-three-level laser oscillator and amplifier. It deals with Gaussian waves, it takes into account the saturation of the pump (which occurs for Yb ion), the stimulated emission at the pump wavelength, the variation of the pump and laser waists along propagation (important for laser diode pumping) and the variation of the laser intensity along propagation. The model is used to predict the output power and the small-signal gain of lasers and amplifiers constituted by the most promising (at the present time) Yb-doped crystals used in the same conditions. This evaluation, established for the CW regime, shows that KYW, KGdW,  $Sc_2O_3$ , S-FAP, C-FAP and YAG have a high potential, depending on the application. Contrary to Deloach *et al.*'s evaluation [11] valid for ns pulse extraction mode, C-FAP and S-FAP crystals no longer have an exceptional position in our evaluation.

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