Yb^{3+}-doped KLu(WO$_4$)$_2$, Nb:RbTiPO$_4$ and KGd(PO$_3$)$_4$ crystals. Growth, characterization and laser operation.

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Abstract

Macrodefect-free samples of Yb^{3+}:KLu(WO$_4$)$_2$, Yb^{3+}:Nb:RbTiOPO$_4$ and Yb^{3+}:KGd(PO$_3$)$_4$ laser crystals were grown using the Top Seeded Solution Growth – Slow Cooling technique. Structural and morphological studies related with the three materials were carried out and discussed. The dispersion of the refractive indices was measured and Sellmeier equations were constructed which are valid in the UV-Vis-IR range. The Stark splitting of the two electronic states of trivalent ytterbium was determined and the CW laser generation was demonstrated in these hosts. This paper shows a review of the main results achieved in FiCMA-FiCNA-URV laboratories in relation with these laser materials in the last years.

Keywords: RbTiOPO$_4$, KGd(PO$_3$)$_4$, KLu(WO$_4$)$_2$, single crystals, Ytterbium solid state lasers.

1.-Introduction

Although the laser effect was discovered in the electronic transitions of Cr^{3+} in a single crystal of Al$_2$O$_3$ [1,2], in recent decades, the development of solid-state lasers was open to other ions and in particular to other lanthanide ions like Yb^{3+} [3-13].

There are many advantages of the Yb^{3+}ion in the development of solid-state lasers, then we will detail some of them. Ytterbium shows a simple electronic structure and has only two electronic states, ^{2}F$_{7/2}$ and ^{2}F$_{5/2}$. Its excited state has no possibility of spectroscopic transfers that can compete with the emission ^{2}F$_{5/2}$→^{2}F$_{7/2}$. Regardless of the host, Ytterbium shows a close to 100% quantum efficiency between absorption and emission (very low quantum defect). The non-radioative transition, which takes place in ytterbium, induces low phonon energy and generates minimal thermal load in the laser material. This minimum thermal load and high lifetime of the above mentioned emission process explains the fact that medium and high power lasers can be developed with this ion.

Yb^{3+} ion can dope a large number of inorganic materials. This circumstance is related with the small ionic radius which show this ion motivated by the lanthanide contraction in any structural coordination. A proof of the possibilities of Yb^{3+}, as active laser ion, is the plenty of papers (more than hundred) published by G. Boulon and co-workers in the last twenty years showing its properties as dopant in more than fifty different materials, both amorphous and crystalline materials. The small radius of Yb^{3+} weakens the screening of the 4f^n shell by 5s and 5p electron shells inducing the enhancement of the crystal field effects on the ytterbium orbitals and producing the broadening of its spectroscopy bands, at least in relation with others lanthanide ions. These wide bands offer the possibility of tuning the laser radiation in a wide range of wavelengths and open the possibility to obtain short or ultrashort pulsed lasers with ytterbium trivalent ions.

But not all are advantages using Yb^{3+} as ion in laser technology, it also shows some inconvenient effect, such as the reabsorption process that takes place with part of the emitted radiation, given the overlapping between their absorption and emission bands. Other disadvantage of Yb^{3+} is that it develops a quasi three levels laser and it is more difficult to achieve population inversion than in four-level scheme which is the case of Nd^{3+} ion. In the case of ytterbium the final laser level in thermally populated and the threshold becomes higher [14,15].

As it was mentioned before, many Yb^{3+} doped crystals were proposed as new laser materials in the last twenty years, and in any case far more than those proposed for other lanthanide ions. In 2008 G. Boulon published a figure-of-merit of Yb^{3+} doped crystals analyzing their spectroscopic data [13]. To find new hosts that improve the performance of ytterbium lasing has been a point of scientific interest in the field of optical materials in the last years. From FiCMA-FiCNA-URV, often in collaboration with G. Boulon and his research group, we have tried to contribute to this scientific target. As a result of this activity, the crystal growth process, spectroscopy characterization and the laser action parameters of three crystals have been determined: Yb^{3+}:KLu(WO$_4$)$_2$ (Yb:KLuW)[16,17], Yb^{3+}:Nb:RbTiOPO$_4$(Yb:Nb:RTP) [18,19,20], Yb^{3+}:KGd(PO$_3$)$_4$ (Yb:KGP) [21,22].

Yb:KLuW is a monoclinic material and its main advantage as laser host is the very high values of the absorption and emission cross sections of Yb^{3+} and other lanthanides, partly due to the strong anisotropy of this biaxial crystal and the possibility to dope it with high concentration of Yb^{3+} without substantial fluorescence quenching because the high distance between the doping ions in the crystalline structure [23].

RTP is a nonlinear optical crystal (noncentrosymmetric) which is possible to dope with Yb^{3+} on the basis of charge compensation by codoping with Nb^{5+} and preserving the value of the effective nonlinearity [24]. This material represents the first realization of laser generation of Yb^{3+} in a crystal member of the KTP family of nonlinear optical

materials. The crystal lases for all three possible polarizations, which guarantees that the lasing action also will be possible along any phase-matching direction of type II self-frequency doubling process.

Yb-KGP is a monoclinic acentric crystalline material. KGP exhibits a broad transmission window extending from about 160 nm to 4 μm which covers the fundamental emission of Yb^{3+} and the spectral range of several harmonics. The rather large band-gap, on one hand, leads to high damage resistivity; on the other hand it allows the use of this host also in the UV range. Another advantage of KGP is its almost isotropic thermal expansion which is important in order to achieve good beam quality in laser operation [21].

In this review paper we present and discuss the main features achieved in the study of these crystals and evaluate their properties. At the same time, we would like to dedicate these pages to honour the scientific contributions of G. Boulon in the field and acknowledge his effort dedicated to the instruction and training of a whole generation of European spectroscopy experts which leads with intelligence and kindness.

2.-Crystal growth of $KLu(WO_4)_2$, $Nb:RbTiOPO_4$ and $KGd(PO_3)_4$ doped with Yb^{3+} ions

Single crystals of KLuW, Nb:RTP and KGP doped with Yb^{3+} ions were grown from high temperature solutions using the Top Seeded Solution Growth-Slow Cooling (TSSG-SC) method. The crystal growth experiments were carried out in cylindrical vertical furnaces and the solutions were prepared in 125 cm^3 cylindrical platinum crucibles.

2.1.- Crystal growth of Yb:KLuW

Potassium rare earth double tungstate single crystals have polymorphic phase transitions and the phase of interest for laser emission is the monoclinic low temperature phase. This phase cannot be obtained directly from the melt and high temperature solutions are usually used for growing it. One of the solvents used for growing Yb:KLuW single crystals is $K_2W_2O_7$ because it has several advantages over other solvents, mainly the absence of foreign ions that could be introduced in the crystal and the low temperature melting of this compound [25].

The solution composition usually used was 12-15 mol % KLuW and 85-88 mol % $K_2W_2O_7$, taking into account the solubility curve [23]. The reagents used were K_2CO_3, Lu_2O_3 (for doping) and WO_3 with purities of 99 %, 99.99 % and 99.5 %, respectively. The solutions, with a weight of 150-200 g, were homogenized for several hours at about 50 K above its saturation temperature. The temperature gradients in the solutions were generally low, around 1 K/cm, with the coldest zone at the center of the solution surface. A KLuW seed with orientation along the *b* crystallographic direction (according previous experiments this is the best orientation to growth high quality

crystals [26]) was located in contact with the center of the solution surface and the saturation temperature was accurately measured. From this temperature, a cooling ramp of 0.1-0.2 K/h for 15-20 K was applied to growth the crystal. During all the growth process, the crystal was rotated at a rate of 40 rpm. Finally the crystal was slowly removed from the solution and maintained at a few millimeters of the solution surface while the furnace was cooled at a rate of 25 K/h. A single crystal just removed from the furnace with the holder used is shown in the inset of Figure 1.a.

The single crystals of Yb:KLuW were transparent and free from inclusions and cracks. As an example, Figure 1.a shows an as-grown Yb:KLuW single crystal. The crystal habit is formed primarily by the {010}, {110}, {$\bar{1}$11} and {310} faces [27]. The composition of the crystals, measured by Electron Probe Microanalysis, leads a calculated coefficient of distribution of Yb^{3+} in these crystals close to unit.

2.2.- Crystal growth of Yb:RTP

Rubidium titanyl phosphate (RTP) melts incongruently and cannot be grown directly from the melt. High temperature solution growth is one of the most used methods to growth these crystals. Two kind of solvents have been used grow RTP single crystals: WO_3 containing solvents (with excess of K_2O and P_2O_5) and self-flux solutions where the solvent is K_2O and P_2O_5 in excess [28]. The doping of RTP with lanthanide ions has been studied at the URV in a previous work [29]. It has been proved that the distribution coefficient of these ions in RTP is very low. The use of codopant elements as Nb or Ta generally increases the lanthanide distribution coefficient in RTP [30].

The growth of $RbTi_{1-x-y}Nb_xYb_yOPO_4$ was made from self-flux solutions. The solution composition was Rb_2O - P_2O_5 - (TiO_2 - Nb_2O_5 - Yb_2O_3) = 42.9 -35.1 - 22 (mol %), where TiO_2 was substituted by Nb_2O_5 and Yb_2O_3 up to 6 and 2 mol %, respectively. The reagents used were Rb_2CO_3 (99%), $NH_4H_2PO_4$ (99%), TiO_2 (99.9%), Nb_2O_5 (99.9%) and Yb_2O_3 (99.99%). The solutions, weighing about 160 g, were homogenized at 50 K above its saturation temperature. The average axial thermal gradient was 1 K/cm, while the radial thermal gradient was 1.8 K/cm, with the cooler point at the center of the solution surface. RTP and Nb:RTP seeds with *c* orientation were used for growing RTP and Yb:Nb:RTP crystals, respectively. In every experiment, one or two seeds located at 1.5 cm from the rotation axis were used and they were fixed to a growth device (see inset of Figure 1.b) which has a turbine dipped in the solution for stirring the solution. The *a* crystallographic direction of the seeds was always in radial direction of the rotating device. In all the experiments the rotation was 65 rpm. After determining the saturation temperature, the solution was cooled at a rate of 0.02-0.1 K/h for 15 K for crystal growth. After removing the crystals from the solution, they were cooled to room temperature at 15 K/h. Figure 1.b shows an as-grown single crystal of Yb:Nb:RTP.

The results show that, when Nb is present as dopant element, the crystal morphology show that the (100) faces are the most developed faces in the crystal and the crystals

take the shape of plates. In addition, these crystals often present cracks starting from the RTP seed, and they can be partially prevented by using Nb:RTP seeds [31]. The concentration of dopants was determined by EPMA. The distribution coefficients of Yb and Nb are far from unity, which means that, during the crystal growth, the Yb and Nb concentration in the solution increases and this tendency was also observed in the crystals [24].

2.3.- Crystal growth of Yb:KGP

Single crystals of Type III $KGd_{1-x}Yb_x(PO_3)_4$ were grown from self-flux solutions. The solution compositions were chosen taking into account the crystallization region of KGdP in the $K_2O-G_2O_3-P_2O_5$ determined previously [32]. Two different solutions were used, $(Gd_2O_3-Yb_2O_3)-K_2O-P_2O_5$ = 4:36:60 (mol %) and $(Gd_2O_3-Yb_2O_3)-K_2O-P_2O_5$ = 6:34:60 (mol %) and the Gd_2O in solution was substituted by Yb_2O_3 up to 15 mol %. The reagents used were Gd_2O_3 (99.99 %), Yb_2O_3 (99.99%) K_2CO_3 (99%) and $NH_4H_2PO_4$ (99%). The solutions, weighing about 200 g, were placed in a region of the furnace with and axial thermal gradient of around 12 K/cm. To improve the solution mixing, an acentric growth device equipped with a platinum turbine submerged in the solution was used. The seed was located in contact with the solution surface at 1.5 cm from the rotation axis, while the turbine was 1.5-2 cm under the seed [33]. A photograph of this growth device is shown in the inset of Figure 1.c [34]. KGP seeds with *a** orientation were used, because in previous studies it has been proved that this orientation ensures a suitable growth rate and avoids the possibility of losing the crystal because of a cleavage plane [22, 33].The rotation velocity of the growth device was kept at 75 rpm with change of the rotation direction every 50 s. The cooling rates used were from 0.05 K/h to 0.1 K/h for 12 K, depending on the ytterbium concentration. It was observed that the number of inclusions in the crystals increased with the increase of the Yb concentration. As an example, Figure 1.c shows a KGP single crystal.

The Yb concentration was measured by EPMA showing that the Yb coefficient of distribution changes from 0.65 to 0.45, with a tendency to decrease with the increase of Yb_2O_3 concentration in the solution [22].

3.-Structural and morphological characterization

3.1.-Yb:KLuW crystal

The crystal structure of $KLu(WO_4)_2$ crystals was determined at room temperature by using single-crystal X-ray diffraction data. They crystallize in the monoclinic system, unit-cell parameters were a = 10.576 (7), b = 10.214 (7), c = 7.487 (2) Å, β = 130.68 (4)°, with Z = 4, in space group *C*2/*c*. The structure is close to that of other monoclinic KREW structures [35-40], and is characterized by a double chain of distorted WO_6 octahedra sharing edges that are aligned along the *c* crystallographic direction through vertex. When compared to other WO_6 octahedra of the family of compounds, those of

KLuW are less distorted, with shorter average W-O distances, and thus with an increased covalent character. Lutetium is eightfold coordinated in this structure by oxygen atoms forming a distorted square antiprism. These polyhedra form a zig-zag single chain along the [101] direction by sharing edges. These square antiprisms are more distorted in KLuW than in the rest of crystals of the monoclinic double tungstates family [35,36]. The Lu-Lu distances within a chain are 4.045 (3) Å, and each lutetium polyhedral chain is surrounded by other four equivalent chains at Lu-Lu distances of 5.982 (3) Å and 6.693 (3) Å, respectively. These distances are of interest when Lu^{3+} is substituted by other active lanthanide ions, and taking into account the energy-transfer phenomena that can occur among lanthanide ions [41-43].

When doped with Yb^{3+}, the *a*, *b* and *c* crystallographic parameters of KLuW increase as the Yb^{3+} concentration increase, while the β parameter remains almost constant, keeping in all cases the monoclinic structure, for Yb^{3+} concentrations up to 50 at.% (even 100 at.% Yb^{3+}, KYbW is also monoclinic). This increase in the unit cell parameters was expected since the ionic radius of ytterbium in this eightfold coordination environment is higher than that of lutetium [44].

Figure 2.a shows the morphology of a KLuW single crystal with an habit formed basically by the {110}, {$\bar{1}$11}, {010} and {310} faces [37]. This morphology matches the one obtained by a periodic bond chains (PBCs) analysis according to the Hartman-Perdok theory [45-47]. Note the importance of the (010) face in the morphology of these crystals, that can be used as an structural reference to cut and polish samples with the appropriate crystallographic orientation for laser applications.

3.2.-Yb:Nb:RTP crystal

$RbTiOPO_4$ crystallizes in the orthorhombic system, with spatial group *Pna*2_1 (point group mm2), being isostructural to the well-known nonlinear optical material $KTiOPO_4$, with unit cell parameters a = 12.974(2) Å, b = 6.494(3) Å and c = 10.564 Å, and Z = 8. The structure is characterized by helicoidal chains of TiO_6 distorted octahedral that are linked between them at two corners and separated by PO_4 tetrahedra [48], with periodic bond chains of -PO_4-TiO_6- along the *a* direction and along the *a*-*c* diagonal. Rb^+ ions are located in the channels left by the network of the TiO_6 octahedra and PO_4 tetrahedra along the *c* direction. The structure contains two inequivalent Ti sites (named Ti(1) and Ti(2)), two inequivalent P sites, two inequivalent Rb sites, and 10 inequivalent oxygen sites. Ti(1) and Ti(2) octahedral alternate along the *c* direction to form helicoidal chains of linked TiO_6 octahedra. Each Ti(1)O_6 octahedron shares two corners with Ti(2)O_6 octahedra through oxygens and the other four corners with PO_4 tetrahedra. Long and short Ti-O bonds alternate in the helicoidal chains, a typical characteristic of the

structure of KTiOPO$_4$ isomorphs. Also, it is observed that the Ti(1)O$_6$ octahedra are larger than the Ti(2)O$_6$ octahedra.

When doped with Nb^{5+} and Yb^{3+}, the unit cell parameters of the structure change. For instance, when doped with Nb^{5+}, parameter *a* became shorter, *b* was slightly longer, and *c* was clearly longer than the corresponding parameters in the RbTiOPO$_4$ structure, while when Yb^{3+} was also introduced in the structure, *a* and *b* parameters were longer and *c* was shorter than those of the RbTiOPO$_4$ crystals doped with Nb^{5+} [49]. Both, Nb^{5+} and Yb^{3+} (or in general a lanthanide ion, Ln^{3+}, such as Er^{3+}) substituted Ti^{4+} in the structure [50]. This explains the changes in the unit cell parameters observed, since Nb^{5+} and Ln^{3+} ions have bigger ionic radius than Ti^{4+} with a 6 fold coordination [44]. The main characteristic of this structure is that the enantiomorphic image of pure RbTiOPO$_4$ is obtained when doping with Nb^{5+}. This means that, while the x and y coordinates for all the atoms did not change very much, the z coordinates shifted considerably and were related to the z coordinates of undoped RbTiOPO$_4$ with a mirror plan at a height of practically a quarter in the *c* direction, similar to what happened in KTiOPO$_4$ crystals doped with Nb^{5+} [51]. Another important characteristic of this structure is that Nb^{5+} and Ln^{3+} ions do not substitute Ti^{4+} ions randomly in the two Ti sites, but we observed that Nb^{5+} ions only occupy Ti(1) sites, while Ln^{3+} ions tend to substitute Ti^{4+} ions preferably in Ti(2) sites [49,50]. The preference of Nb^{5+} to occupy only Ti(1) sites seems to have an electrostatic origin, since the Ti-Rb bond distances in this position are longer than those in the Ti(2) site, implying that the electronic repulsion in the Ti(2) position is higher than that in the Ti(1) site. Consequently, Nb^{5+} tends to occupy Ti(1) positions to avoid this electrostatic repulsion. Furthermore, we observed a tendency for Yb^{3+} ions to preferentially occupy sites close to Nb^{5+} ions, forming Nb^{5+}-Yb^{3+} pairs in the structure, supporting the fact that Yb^{3+} tend to occupy Ti(2) sites [52].

Figure 2.b shows the typical morphology of a Yb:Nb:RTP crystal, comprising the {100}, {201}, {110} and {011} faces [53]. When Nb^{5+} is present, the {100} face tends to be the most developed face in the crystals.

3.3.-Yb:KGP crystal

Type-III KGd(PO$_3$)$_4$ (KGP) crystallizes in the monoclinic system, with *P*2$_1$ spatial group [54], being isostructural to KNd(PO$_3$)$_4$ [55]. The unit cell parameters for KGP are a = 7.255(4) Å, b = 8.356(5) Å, c = 7.934(5) Å, and β = 91.68(5)°, with Z = 2. One structural position for Gd and K, four positions for P, and twelve inequivalent positions of O can be found in this structure. All these atomic positions are doubled by the 2-fold screw axis parallel to the [010] crystallographic direction appearing in the spatial group of symmetry. The P atoms are linked to the nearest oxygen atoms forming slightly distorted PO$_4$ tetrahedra. These tetrahedra form chains sharing two of their vertexes, which is a characteristic of the phosphates type III structure [56]. The basic unit of these chains is made of four PO$_4$ tetrahedra. This basic unit is repeated by periodicity to

generate zigzag PO_4 chains along the ***a***-crystallographic direction. Gd atoms are linked to eight oxygen atoms to form distorted dodecahedra. These GdO_8 polyhedra share O atoms only with the PO_4, which implies that they form isolated dodecahedra in the structure. The K^+ ions are also eight-coordinated, exhibiting the largest bond distances in the structure, so they are weakly bonded in the crystal lattice. It is also worthy to mention that Gd^{3+} and K^+ atoms alternate with a zigzag arrangement parallel to the PO_4 chains.

It was difficult to incorporate high concentrations of Yb^{3+} in the structure of KGP without changing its crystal structure. In fact, at high Yb^{3+} concentrations, the material tended to crystallize also in the monoclinic system but with the centrosymmetric $C2/c$ space group with cyclical geometry [22] instead of the desired $P2_1$ group [54], despite that the dopant acceptance seemed not to be limited by the ionic radius of Yb^{3+}. The crystallization of the $C2/c$ phase depends on the $(Yb_2O_3 + Gd_2O_3)$ / K_2O molar ratio in which the Yb:KGP crystals have been grown and the doping concentration of Yb^{3+} [21,22]. For K_2O molar concentrations above 85 % the $P2_1$ phase crystallizes for Yb^{3+} concentrations as high as 50 at.%, but the $C2/c$ phase was find together with the $P2_1$ phase when the K_2O molar ratio was 85 % or lower, even for Yb^{3+} concentrations as low as 0.5 %.

The crystal cell parameters ***a***, ***b***, ***c*** and V slightly decreased in a linear way as the concentration of Yb^{3+} increased in crystals, indicating that it progressively substituted gadolinium in its structural position [22].

Figure 2.c shows the typical morphology of a KGP crystal, with an habit formed by the {001}, {100}, {011}, {0$\bar{1}$1}, {110}, {1$\bar{1}$0}, {10$\bar{1}$}, {101}, {1$\bar{1}\bar{1}$} and {111} [32]. Despite many of these faces are not equivalent taking the account the symmetry group, in final morphology of the crystal they tend to be very similar because the β angle is close to 90º and the unit cell parameters have a similar value. This implies that for the correct face indexation and crystallographic orientation of the samples for laser applications it is necessary to use X-ray diffraction techniques.

4.-Optical properties

The absorption spectra of KLuW, Nb:RTP and KGP were recorded using a UV-VIS and a FTIR spectrometer, depending on the spectroscopic range. Figure 3 shows that the transparency window of these hosts extends from the UV to the IR [21,27,37,57]. For example, KGP has an UV edge at zero transmission at 160 nm and its optical transparency extends up to 4 µm, KLuW shows a flat transparency window from 365-5110 nm, with a UV cut-off wavelength near 300 nm. Nb:RTP has a $\lambda_{cut-off}$ of 340 nm and its transparency extents up to 4500 nm. The broad transparency window of these materials is a key point for using these materials as a crystalline matrix for laser active ions emiting in a wide region of the spectrum. Also, combining the low $\lambda_{cut-off}$ in the UV of KGP and RTP with the nonlinear character of these crystals, will allow to use these crystals to produce deep UV laser radiation from high order harmonics, especially in the case of KGP.

The knowledge of principal refractive indices in the whole transparency window of the crystal is important for designing the laser operation of the crystal. Also, for nonlinear optical materials, the dispersion equations will allow to determine the phase-matching conditions for sum or difference frequency conversion. The monoclinic phases of KLuW and KGP are biaxial crystals and the orientation of the dielectric tensor is required. In monoclinic crystals, one of the principal optical axes (in this case N_p) coincides with the **b** crystallographic axis and the other two principal axes, N_g and N_m, are in the **a-c** plane. The optical frames of KGP and KLuW were oriented by using two crossed Glan-Taylor polarizers. Using a He-Ne laser (λ=632.8 nm), we found a minimum transmission when the KLuW sample, with **b** positive pointing toward the reader, was rotated 18.5° from the **c** direction (N_g) and 58.7° from the **a** direction (N_m), both in the clockwise direction. For KGP, these angles are, respectively, 37.3° and 39.1° (Figure 4). RTP crystallizes in the orthorhombic crystal system, and then, the crystallographic axes coincide with the orthonormal dielectric frame.

The mininum deviation method was used to determine the dispersion of the three refractive indices. Two prisms, cut in different principal planes, were used with an accuracy of $5 \cdot 10^{-4}$. The best fit of all the recorded data leads to the determination of three Sellmeier dispersion equations, including one UV pole and one IR correction term

Figure 5 gives the dependency of the principal refractive indices with the wavelength for KLuW, RTP and KGP and Table 1 listes the four parameters describing the dispersion of each material in each principal direction.

5.-Yb^{3+} spectroscopy in KLuW, Nb:RTP and KGP

Yb^{3+} has a 4f^{13} ground state configuration consists of two $^{2s+1}L_J$ manifolds, the $^2F_{7/2}$ ground state and the $^2F_{5/2}$ excited state, which usually is around 10000-11000 cm^{-1}. In the three different hosts described in the present paper, the Yb^{3+} substitutes the Lu^{3+}, the Ti^{4+}(Ti(2)) and the Gd^{3+} unique site, respectively for KLuW, Nb:RTP and KGP. The symmetry sites are C$_2$ for the KLuW and for the two other hosts and C$_1$. The electrostatic crystal field effect (Stark effect) breaks partially the (2J+1) degeneracy of the ion levels in the host, splitting in Krammers doublet sublevels of (2J+1)/2. So, typically, the ground state of Yb^{3+} is split in 5 sublevels and the excited state in 3 sublevels. Table 2 summarizes the energy values of the sublevels of Yb^{3+} in the three different hosts.

From the crystal splitting, it is already possible to affirm that the crystal field strength follows the tendency RTP>KLuW>KGP, being in the RTP the largest one. This is highly correlated with the degree of distortion of the coordination polyhedral of the Yb^{3+} in the host and also the first probable Yb^{3+}-O distances in the crystalline structure, reported in Table 3.

As the number of unpaired 4f electrons of Yb^{3+} configuration is odd, and with the local symmetries described above, no polarization selection rules are expected for the transitions. However, the anisotropy of the host induces some changes in the absorption in relation with the polarization of the incident light. Ytterbium spectroscopy has been performed in polarized way, using the principal optical axes (dielectric frame) as a frame. Observing the polarized absorption of these hosts at room temperature in Figure 6; it can be highlighted that the lowest anisotropy could be assigned to the KGP host, but in the case of Nb:RTP and KLuW, the anisotropy is large.

The values of the maxima absorption cross section are summarized in table 4. The host dependence of the absorption cross section should be related with the refractive index values and also, the Judd-Ofelt values, which are directly related with the strength of the oscillator for the electronic transition [59,60].

The reciprocity method was used to compute the emission cross sections from the absorption cross sections measured at room temperature and the determined sublevel energy values. Figure 7 shows the calculated emission cross-section for the three different hosts in the polarization mainly used in the laser action.

Lifetime of $^2F_{5/2}$ emitting level in ytterbium is related to a radiative emission with branching ratio equal to unity. The reabsorption and the total internal reflection effects will affect the experimental measured decay times, elongating their value.

The fluorescence decay time was measured by the pinhole method which avoids radiation trapping in KGP and KLuW crystals. For KGP, the extrapolated for zero diameter result, at room temperature, was 1.22 ms but the deviation from this value for all the pinhole diameters used, between 0.6 and 2.1 mm, was only ±1%. Thus,

reabsorption was negligible in this crystal. For KLuW, the measurements by pinhole method lead to a lifetime value measured of 299 μs. Finally, lifetime measurement of Yb in RTP, without the pinhole correction, lead to a value of τ_{emi}= 2.2 ms. These values repeat the tendency of the emission cross section; as larger is the probability of the radiative emission, shorter lifetime is obtained. The radiate lifetimes reported in Table 4, were calculated using Füchtbauer-Ladenburg equation or Weber approximation.

6.-CW laser operation results in Yb:KLuW, Yb:Nb:RTP, and Yb:KGP

The laser performance of the three Yb-doped oxide crystals (KLuW, Nb:RTP and KGP) grown at FiCMA-FiCNA laboratories have been studied under Ti:sapphire laser pumping in continuous-wave (CW) regime. The laser set-up used to achieve lasing is the same in the three cases.

The pump beam from a home-made Ti:sapphire laser (FWHM<1 nm, max. 2.0 W) was focused onto the crystals by a 6.28 cm lens L to a Gaussian waist of ≈30 μm. It has been tuned to the maximum of absorption for each crystal. The ≈150 cm long, astigmatically compensated cavity consisted of two folding mirrors (M1 and M2), both with radius of curvature equal to -10 cm, a rear plane mirror, M3, and the plane output coupler, M4, with transmission of 1%, 3% or 5%. The laser set-up is shown in Figure 8.

The samples were inserted under Brewster angle between the two folding mirrors (M1 and M2) and no special cooling was applied.

6.1.- Laser operation of Yb^{3+} in KLuW

For laser generation, Yb:KLuW samples grown from 5 and 10 mol % Yb-content in the solution were used. The concentrations of Yb-ions in the crystals were 4.30×10^{20} and 8.35×10^{20} cm^{-3}, respectively and the thicknesses 2.8 and 2.2 mm, respectively. All samples were cut and polished with their parallel faces normal to the N_p-principal optic axis.

With T_{OC}=2.8% the Yb:KLuW laser threshold could be reached for 965 nm < λ_P < 1005 nm. The optimum Ti:sapphire laser pump wavelength for the 5% Yb-doped KLuW sample was λ_P=982 nm and for the 10% Yb-doped KLuW sample was λ_P=979 nm. On the basis of the absorption spectrum, the necessity to pump below the main absorption maximum in the case of higher doping can be explained by the lower absorption there which allows more homogeneous pumping along the sample thickness with reduced reabsorption. The results obtained at the optimum λ_P with the two Yb:KLuW samples are shown in Figure 9 for two values of T_{OC}: 2.8% (Figures 9.a and 9.c) and 5% (Figures 9.b and 9.d). The longer generation wavelengths λ_L with the higher doped KLuW are a consequence of the increased reabsorption. Since the saturated gain always equals the total losses, the decreasing λ_L when increasing T_{OC} can be also attributed to the wavelength dependent reabsorption losses. The most important conclusion from the comparison of the 5% and 10% Yb-doped KLuW samples is that

the crystal quality and presumably the upper level lifetime remain unaffected by the increased dopant density. This is clear from the fact that the results in Figure 9, having in mind the slightly different wavelengths and absorbed powers, are almost identical in terms of conversion efficiency and threshold.

CW lasing in Yb:KLuW was carried out and conversion efficiencies as high as 50% and output powers of the order of 1 W without active cooling the crystal were achieved. Diode pumping at 980 nm, as well as femtosecond laser operation with this crystal has been demonstrated in other works [61,62] showing excellent results.

6.2.- Laser operation of Yb^{3+} in Nb:RTP

The Yb:Nb:RTP sample used had dimensions of ≈3 mm along the *a*- and *c*- axes, and ≈2.5 mm along the *b*-axis, and four of its faces (⊥ *a* and ⊥ *b*) were polished. The Yb-density in the crystal was 1.87×10^{20} cm^{-3}.

The input-output characteristics of the Yb:Nb:RTP laser are summarized in Figure 10 for the three possible polarizations and propagation along the *a*-axis (Figure 10.a) and *b*-axis (Figure 10.b). The pump wavelength was 972.1 nm (*E*//*a* and *E*//*c*) and 972.7 nm (*E*//*b*). The highest absorption cross section is for *E*//*b* with maximum absorption for propagation along the *a*-axis. In this case, a maximum output power of 154 mW at 1050.6 nm was obtained using an output coupler with *T*=1%, for an absorbed pump power of 386 mW. The corresponding slope efficiency was *η*=42.5% and the threshold (absorbed power) was P_{th}=35 mW.

The performance of the laser for *E*//*c* and propagation direction along the *a*- and *b*- axes was very similar although the sample thickness was different. For propagation along the *b*-axis, the slope efficiency reached *η*=60.1% for *T*=3% while propagation along the *a*-axis, resulted in an output power of 115 mW with *T*=1%, for an absorbed pump power of 323 mW. For *E*//*a* and propagation along the *b*-axis, laser generation was obtained only in the case *T*=1% and the slope efficiency was lower compared to *E*//*b* and *E*//*c*, presumably because of the smaller gain cross-section. The laser wavelength was basically the same for the three polarizations and the two output couplers, i.e. the maximum of the gain curve remains unchanged. The extremely low laser thresholds are related to the large splitting of the ground state, as mentioned above. With lasing interrupted, the absorption of the sample was bleached at the maximum incident pump power of 1.8 W. Under lasing conditions the absorption was still quite low (13% for *E*//*a*, 25-20% for *E*//*b*, and 15-20% for *E*//*c* depending on the output coupler used) and remained almost constant with increasing pump power. This is a consequence of the low Yb-ion density in the crystal and the relatively low absorption cross-sections. However, in the lasing state, the intracavity power in the three-level Yb-system increases the pump saturation intensity and this balances the bleaching effect.

Continuous-wave laser operation of Yb at room temperature in the orthorhombic Nb:RTP crystal was achieved around 1050 nm for all three possible polarizations. Further increase of the doping level could drastically improve these initial laser results with Yb:Nb:RTP in terms of output power. Diode-pumping and further power scaling

should be also possible having in mind the broad absorption spectrum of Yb:Nb:RTP. The extremely broad tunability achieved is also promising for mode-locking to obtain femtosecond pulses and self-frequency conversion with this material.

6.3.- Laser operation of Yb^{3+} in KGP

The Yb:KGP sample used had dimensions of 2.34, 2.68 and 2.47 mm along the N_p, N_m, and N_g principal optical axes. The Yb-density in the crystal was 1.01×10^{20} cm^{-3}.

Continuous-wave laser operation was obtained for the first time in this monoclinic material at room temperature for pumping with $E//N_m$ (propagation along N_g) and with $E//N_p$ (propagation again along N_g). The laser always had the same polarization as the pump due to the Brewster orientation. No generation was possible for pumping with $E//N_g$ although it was tried this for propagation both along N_m and along N_p.

The input-output characteristics obtained for the two polarizations are shown in Figure 11 against the absorbed pump power for two output couplers (T_{oc}=1 and 3%). The relevant laser parameters (slope efficiency η with respect to the absorbed power, oscillation wavelength λ_L and threshold) are summarized in Table 5.

With a maximum incident pump power of roughly 2 W (corresponding to an absorbed power of 206 mW at 977.1 nm), the maximum output power for $E//N_m$ was 93 mW (T_{oc}=1%). The corresponding slope efficiency was η=53.2%. Pumping with polarization parallel to N_p, the maximum output power reached 72 mW for an absorbed power of 294 mW, also with T_{oc}=1%. In this case the slope efficiency was lower, η=31.3%. The laser thresholds for $E//N_m$ and $E//N_p$ were 51 and 74 mW, respectively, both for T_{oc}=1%. The lower slope efficiency and higher threshold for $E//N_p$ can be explained by the lower gain.

For T_{oc}=5% the output power reached 12 mW (only for $E//N_m$) and reliable estimation of the slope efficiency was not possible. The oscillation wavelength was as short as 1012.1 nm in this case. The shorter wavelength at higher output coupler transmission is typical for Yb-lasers and is related to the maximum of the gain curve. In this case, there's the oscillation on the $^2F_{5/2}(0')\rightarrow ^2F_{7/2}(2)$ transition.

Under lasing conditions the absorption of the sample was quite low (not more than 15%) but almost constant. This is a consequence of the low Yb-ion density in the crystal and the relatively low absorption cross-sections. Under non-lasing conditions the absorption was completely bleached at the maximum incident power. However, in the lasing state, the intracavity power in the three-level system of ytterbium increases the saturation intensity for the pump and this balances the bleaching effect.

Although the maximum output power achieved (93 mW) was limited by the available size and doping level of the crystal, the more than 55% slope efficiency obtained with this first sample is rather promising for the future. Also diode-pumping is planned for the future.

Conclusions

Ytterbium doped KLuW, Nb:RTP and KGP single crystals were grown using Top-Seeded Solution Growth - Slow Cooling method. Structural, morphological, optical and spectroscopical characterization of these materials was reviewed. Summarized CW laser parameters of ytterbium in these hosts were reported in the present paper. CW lasing in Yb:KLuW was carried out with conversion efficiencies as high as 50 % and output powers of the order of 1 W. In Yb:Nb:RTP samples, with a pumping wavelength of 972.7 nm, the highest absorption cross section was for $E//b$ with maximum absorption for propagation along the a-axis. A maximum output power of 154 mW at 1050.6 nm was obtained with a slope efficiency of 42.5 %. For $E//c$ and propagation direction along the b-axis, the output power was 115 mW and the slope efficiency was 60.1% for propagation along the a-axis. The extremely broad tunability achieved is also promising for mode-locking to obtain femtosecond pulses and self-frequency conversion with this material. CW laser operation at room temperature in a Yb:KGP for $E//N_m$ resulted in a maximum output power of 93 mW, with a slope efficiency of 53.2 %. Current advances in these hosts doped with Yb^{3+} are pulsed Yb^{3+} lasers based on KLuW single crystals and the use of the nonlinear optical properties of Yb;Nb:RTP and Yb:KGP combined with laser action of Yb^{3+} and the fabrication of waveguide platforms. In addition, the low UV cut-off of the transparency window of KGP offers a great potential for deep UV uses.

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Table 1 Sellmeier coefficients of KLuW, RTP and KGP.

Principal Refractiveindex	A_i	B_i	C_i (μm^2)	D_i (μm^{-2})
n_g-KLuW	3,5833	0,7351	0,2670	0,0295
n_m-KLuW	3,3699	0,7431	0,2619	0,0433
n_p-KLuW	3,2175	0,7538	0,2507	0,0508
n_x-RTP	1,6795	1,4281	0,0325	0,0119
n_y-RTP	2,0360	1,0883	0,0437	0,0090
n_z-RTP	2,2864	1,1280	0,0562	0,0188
n_g-KGP	1,7728	0,7782	0,1391	0,0091
n_m-KGP	1,7624	0,7667	0,1304	0,0193
n_p-KGP	1,7404	0,7479	0,1374	0,0138

Table 2. Energy values in cm^{-1} of the Yb^{3+} sublevels in the three hosts.

hosts		KLuW	Nb:RTP	KGP
$^2F_{7/2}$	(0)	0	0	0
	(1)	175	309	125
	(2)	435	521	298
	(3)	559	956	382
$^2F_{5/2}$	(0')	10187	10289	10245
	(1')	10498	10471	10309
	(2')	10735	11069	10559
[ref]		[27]	[58]	[21]

Table 3.Geometrical parameters of the Yb^{3+} structural environment in the three different hosts.

Sample	KLuW	Nb:RTP	KGP
Polyhedral distortion	4.3(5)	22	0.06
M-O distance [Å] (M=Lu, Ti and Gd)	2.385	1.9	2.406
[ref]	[37]	[49,52]	[54]

Table 4. Yb^{3+} spectroscopic parameters in the three different hosts

	KLuW	Nb:RTP	KGP
$[Yb^{3+}]$ ions/cm³, % at. Yb substitution	$0.45 \times 10^{20}, 0.5\%$	$1.87 \times 10^{20}, 2.1\%$	$1.01 \times 10^{20}, 2.4\%$
Maxima $\sigma_{abs}[10^{-20}]$at/cm², FHWM [nm], λ [nm], pol	$11.8, 3.6, 981.1, N_m$	$0.77, 4.0, 972.3, b$	$1.17, 17.0, 977, N_m$
σ_{emi} [x 10^{20}]at/cm², FHWM[nm], λ_L[nm], pol	$1.01, 22, 1040, N_m$	$0.27, 9, 1049.4, c$	$0.56, -, 1014, N_m$
τ_{rad}[ms], τ_{emi}[ms]	0.320, 0.299	2.7, 2.2	1.57, 1.22
[ref]	[23,27]	[58]	[21]

Table 5. Slope efficiency (η), laser wavelength (λ_L), and threshold of the KYb$_{0.024}$Gd$_{0.976}$(PO$_3$)$_4$ laser in dependence on the output coupler (T_{oc}) used

T_{oc}[%]	η [%]		λ_L[nm]		Threshold [mW]	
	$E//N_m$	$E//N_p$	$E//N_m$	$E//N_p$	$E//N_m$	$E//N_p$
1	53.2	31.3	1017.1	1016.3	51	73.8
3	55.6	-	1013.8	1013.5	70.5	127.8
5	-	no lasing	1012.1	no lasing	131.8	no lasing

Figure caption list:

Figure 1.- a) Yb:KLuW crystal and holder detail, b) Yb:Nb:RTP crystal and holder detail c) KGP and holder detail.

Figure 2. Crystal morphology of a) KLuW, b) Nb:RTP and c) KGP single crystals.

Figure 3. a) Unpolarized transmission of KLuW, Nb:RTP, and KGP from the UV to mid-IR b) Zoom of the transparency window in the UV edge.

Figure 4.- Optical ellipsoid for a) KLuW and b) KGP for λ=632.8 nm at room temperature

Figure 5.- Dispersion of the principal refractive indices of KLuW, Nb:RTP and KGP.

Figure 6.- Polarized optical absorption cross section at room temperature of Yb^{3+} ion, corresponding to the electronic transition $^2F_{7/2}{\to}^2F_{5/2}$ in the three different hosts. a) KLuW; b) Nb:RTP and c) KGP.

Figure 7.- Polarized optical emission cross section at room temperature of Yb^{3+} ion, corresponding to the electronic transition $^2F_{5/2}{\to}^2F_{7/2}$ in the three different hosts.

Figure 8.- Experimental set-up for the laser characterization of the three crystals.

Figure 9.- Output power P_{out} versus absorbed power P_{abs} in double pass (symbols) obtained at the corresponding optimum λ_P, and linear fits (lines) for calculation of the slope efficiency η for the 5% (a, b) and 10% (c, d) Yb-doped KLuW samples and two different output couplings (T_{OC}). The wavelength λ_L is indicated both at threshold (bottom values) and at maximum P_{out} (top values).

Figure 10.- Input-output characteristics of the Yb:Nb:RTP laser for different output coupling (T) and polarization (E): Pth: threshold absorbed power, η: slope efficiency, λ: oscillation wavelength.

Figure 11.- Room temperature continuous-wave laser performances of a) Yb:KGP for *E//N_m* and b) *E//N_p*. solid lines are fits to the experimental points for estimation of the slope efficiency.