Quantum Electronics Letters

CW Laser Operation of $KLu_{0.945}Tm_{0.055}(WO_4)_2$–$KLu(WO_4)_2$ Epilayers Near 2 μm

Òscar Silvestre, Maria Cinta Pujol, Magdalena Aguiló, Francesc Díaz, Xavier Mateos, Valentin Petrov, and Uwe Griebner

***Abstract*—Thin epilayers of monoclinic $KLu(WO_4)_2$ doped with thulium are grown by liquid phase epitaxy on undoped $KLu(WO_4)_2$ substrates. The epitaxial composites are optically characterized and implemented in a cavity, longitudinally pumped (normal to the layer) by a Ti:sapphire laser. Tunable (1894–2039 nm) continuous-wave laser operation is achieved with maximum slope efficiency as high as 64% with respect to the absorbed pump power at 802 nm. The output power is limited by the crystal absorption.**

***Index Terms*—Crystals, double tungstates, epitaxial layers, lasers, rare earth compounds, solid lasers, thulium.**

I. INTRODUCTION

THE $^3F_4 \rightarrow {}^3H_6$ transition of Tm^{3+} is very promising for tunable laser emission in the 2-μm spectral range. Such solid state lasers can be pumped by the widely spread AlGaAs diodes emitting in the 800-nm spectral range which were developed for Nd-lasers. Among the thulium doped materials, monoclinic $KRE(WO_4)_2$ tungstates (RE =Y, Gd and Lu; hereafter KREW) are distinguished by the largest cross sections, partly due to the strong anisotropy, the higher doping levels possible, and the Stimulated Raman Scattering (SRS) activity [1]. Recently, continuous-wave (CW) laser operation on the $^3F_4 \rightarrow {}^3H_6$ transition was achieved in Tm-doped KYW [2], KGdW [3], and KLuW [4]. With Tm:KLuW, the maximum output power was 4 W and the slope efficiency reached 69% while the tunability extended from 1800 to 1987 [4].

The possibility to increase the doping level together with the high absorption cross sections is very interesting for the thin-disk laser concept which can be used to scale the output power by optimizing the one-dimensional longitudinal heat flow [5]. The heat removal efficiency scales inversely with the thickness of the active element. Thin-disk geometry has been realized so far only with one Tm host, bulk YAG (0.65-mm-thick sample with 6% Tm-doping), which provided a CW output power as high as 4 W [6]. Using KREW hosts, it is in principle possible to achieve such large absorption coefficients that the appropriate active element thickness would be below the limit set by the processing requirements and the opto-mechanical properties (less that 0.1 mm). However, we recently demonstrated that liquid phase epitaxy (LPE) can be used to grow thin single crystalline layers of Yb-doped monoclinic double tungstates on undoped material. In the case of this dopant the best results were achieved with KLuW used as a host and substrate [7]. The close ionic radii of Lu and Tm are a good prerequisite also for the growth of defect-free epitaxial layers of Tm-doped KLuW on undoped KLuW. In the present paper we report on the growth and characterization of such epitaxial composites and demonstrate, for the first time to our knowledge, efficient laser operation and tunability in the 2-μm spectral range pumping normal to the active layer.

II. GROWTH OF Tm:KLuW–KLuW EPITAXIAL COMPOSITES

KLuW is a monoclinic crystal with $C2/c$ space group symmetry. The unit cell parameters are $a = 10.576(7)$ Å, $b = 10.214(7)$ Å, $c = 7.487(2)$ Å, and $\beta = 130.68°(2)$. More information on its structural and other physical properties can be found in [8]. Single crystals of KLuW to be used as a substrate were grown by the top seeded solution-growth slow-cooling (TSSG-SC) method using $K_2W_2O_7$ as a solvent. A detailed description of the growth procedure can be found elsewhere [8]–[10]. The solution composition was 12 mol. % KLuW–88 mol. % $K_2W_2O_7$ and the solution weight was around 200 g. The axial temperature gradient in the solution was about 0.1 K/mm, with the bottom hotter than the surface. The crystals were grown on ***b*** oriented seeds perpendicular to the surface of the solution, located at the center of the crucible. The cooling rate was 0.2 K/h for 20 K. The average growth rate was 0.059 g/h. The crystals were rotated at a constant velocity of 40 rpm. At the end of the growth process they were removed slowly from the solution to avoid thermal shock and located slightly above the solution surface, then cooled at a rate of 25 K/h down to room temperature.

Manuscript received August 15, 2006; revised October 24, 2006. This work was supported by the EU-Commission under Project DT-CRYS STRP-505580-1, in part by the Generalitat de Catalunya under the Project 2005SGR658, and in part by the MEC (Ministerio de Educación y Ciencia of the Spanish government) under Projects MAT-05-06354-C03-02, MAT-04-20471-E and CIT-020400-2005-14. The work of X. Mateos was supported in part by the Secretaria de Estado de Educacion y Universidades of Spain and from the Fondo Social Europeo.

Ò. Silvestre, M. C. Pujol, M. Aguiló, and F. Díaz are with the Group of Física i Cristal·lografia de Materials (FiCMA), Universitat Rovira i Virgili, E-43007 Tarragona, Spain (e-mail: oscar.silvestre@urv.cat; mariacinta.pujol@urv.cat; magdalena.aguilo@urv.net; f.diaz@urv.net).

X. Mateos, V. Petrov, and U. Griebner are with the Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, D-12489 Berlin, Germany (e-mail: mateos@mbi-berlin.de; petrov@mbi-berlin.de; griebner@mbi-berlin.de).

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/JQE.2006.890383

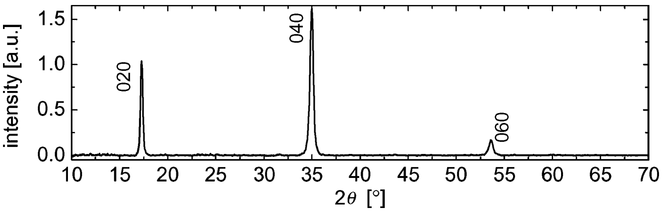

Fig. 1. 2θ-scan X-ray diffraction pattern of the epitaxial layer.

The LPE growth experiments were carried out in a special vertical furnace built with a wide axial zone of uniform temperature so that the vertical temperature gradient in the solution is practically zero in the zone of the epitaxial growth which is important for the homogeneity of the epitaxial layer. The crucibles used were cylindrical, 30 mm in diameter and 40 mm high, filled with about 70 g of solution.

The optimum composition of the solution is determined as a compromise between the growth rate and the quality of the interface obtained. Taking into account the solubility curve of KLuW in $K_2W_2O_7$, the composition of the solution was chosen to be 7 mol. % $KLu_{0.95}Tm_{0.05}(WO_4)_2$–93 mol. % $K_2W_2O_7$. The lower percentage of the solute, the same as the one used in the case of Yb-doped layers [7], in comparison to the bulk crystal growth permits better control of the growth rate according to the solubility curve [11]. The Tm doping level was also chosen as a compromise, on one hand to ensure good optical quality of the composite having smaller lattice mismatch, taking into account our experience with the bulk Tm:KLuW crystals [4], and on the other hand aiming at sufficiently high concentration in order to have highly absorbing layers whose thickness is low enough so that it is otherwise impractical for free-standing active elements. The reagents were the same as in the bulk substrate growth and Tm_2O_3 was used for substitution. The molar ratios were: 32.16% K_2CO_3, 1.11% Lu_2O_3, 0.06% Tm_2O_3, and 66.67% WO_3, in order to obtain the stoichiometry specified above for the solution.

Once the solution had been homogenized, its saturation temperature was accurately measured with a ***b***-oriented KLuW seed rotating at 40 rpm. The growth and dissolution rates were determined with the seed tied to an alumina rod by means of a mechanical micrometric comparator with a precision of ±0.01 mm. The saturation temperature T_s is 1138–1140 K.

The substrate plates were prepared by cutting and polishing 2-mm-thick slices oriented perpendicular to the ***b*** crystallographic direction. In analogy with our previous experience with LPE grown Yb-doped epitaxial composites [11], the best optical quality (best flatness and lowest density of micromorphologies) and the fastest growth rate were achieved for the (010) face of the epitaxial films, i.e., along the $\boldsymbol{b}//\boldsymbol{N_p}$ direction which allows interaction with light polarized with $\boldsymbol{E}//\boldsymbol{N_m}$ ensuring maximized interaction cross sections. $\boldsymbol{N_p}$ and $\boldsymbol{N_m}$ denote two of the three orthogonal principal optical axes defined from the relation $n_p < n_m < n_g$ for the refractive indices. First, the substrates were carefully cleaned by immersion, with rotation, in HNO_3–$H_2O = 1/1$ (5 min), distilled water (5 min), acetone (5 min), and ethanol (5 min). Then they were slowly inserted into the furnace to prevent thermal stress and heated for about one hour above the surface of the solution. Finally they were dipped into the solution with their ***c***-axis perpendicular to the surface of the solution. The epitaxial growth took place for several hours with the crystal rotating at 40 rpm, at 6 K below T_s. The supersaturation, achieved at temperatures lower than T_s, is the main driving force for the growth process and it greatly affects the quality of the epitaxial film. The 6 K difference corresponds to a supersaturation coefficient, σ, around 5.3% where $\sigma = 100(c/c_0 - 1)$, and c and c_0 are the solute concentration at the growth and saturation temperatures, respectively, determined from the solubility curves of KLuW in the $K_2W_2O_7$ solvent [11]. The crystalline composites were removed from the flux and cooled slowly to room temperature to avoid thermal stresses.

As can be seen from the X-ray diffraction pattern shown in Fig. 1, the 2θ-scan of the layer, similar to that of the substrate, exhibits only the (0k0) reflections. The FWHM of the (020) peak is 0.18° and 0.16° for the layer and the substrate, respectively. This is indicative of a ***b***-oriented epitaxy. The obtained Tm:KLuW epitaxial layers were transparent, colorless, with single crystalline quality, and free of macroscopic defects, which indicates that the established growth process is almost layer-by-layer, leading to a quasi-flat surface. Nevertheless, for optical characterization and laser applications it was necessary to polish the samples. In the process of polishing, the thickness was controlled by means of a mechanical micrometric comparator with accuracy ±1 μm.

For the laser experiments, an epitaxial composite of $KLu_{0.945}Tm_{0.055}W$–KLuW (010) was polished to high optical quality. The final sample size was $3.4 \times 3 \times 1.7$ mm^3 $(\boldsymbol{N_m} \times \boldsymbol{N_g} \times \boldsymbol{N_p})$. Using a SENSOFAR PL$\mu$ 2300 confocal microscope, we were able to inspect the quality of the layer surface after polishing. The surface was flat over areas sufficiently large for longitudinal pumping, normal to the layer. Fig. 2(a) shows a photograph of the sample and Fig. 2(b)—a plot of the profile of the (010) epitaxial surface. The surface radius of curvature is 2.5 and 1.5 m along the $\boldsymbol{N_g}$ and $\boldsymbol{N_m}$ directions, respectively, and the roughness has a rms value of 20 nm. The final thickness of the epitaxial films was measured by optical microscope. From the inset of Fig. 3, it can be seen that the substrate-film interface is clearly distinguishable and the film thickness can be measured with the magnification of the microscope. The obtained layer thickness is 130 μm.

The actual composition of the doped layer was derived from electron probe microanalysis with wavelength dispersive spectroscopy (EPMA-WDS) using a Cameca SX 50 equipment. Moreover, measuring the Tm concentration along the interface, it was possible to determine the doping ion diffusion into the substrate. Fig. 3 shows the distribution of the Tm^{3+} concentration in direction normal to the epitaxial film; it can be observed that the width of the interface does not exceed 2 μm. The composition of the epitaxial layer is $KLu_{0.945}Tm_{0.055}W$. Hence, the distribution coefficient of thulium is $K = 1.10$.

III. LASER OPERATION OF Tm:KLuW–KLuW

The laser setup used in the present work (Fig. 4) was similar to that described in [4]. The astigmatically compensated X-type cavity had a total length of 90 cm. Output couplers (M4 in the

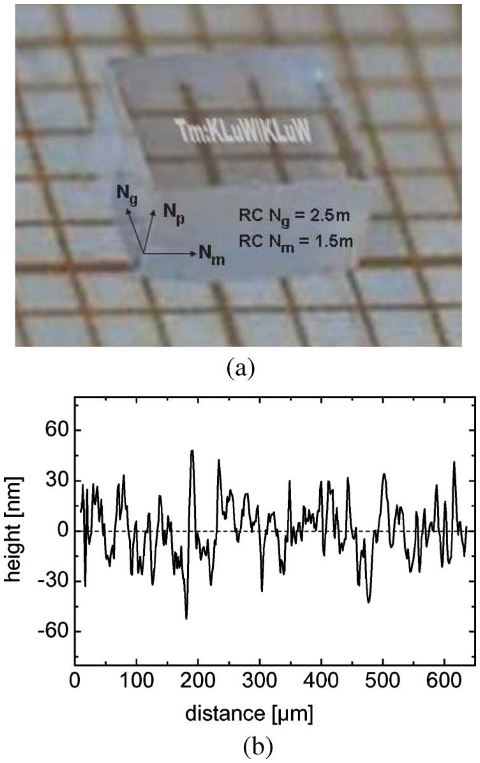

Fig. 2. (a) Photograph of the Tm:KLuW–KLuW epitaxial sample used in the laser and (b) roughness profile of its layer surface.

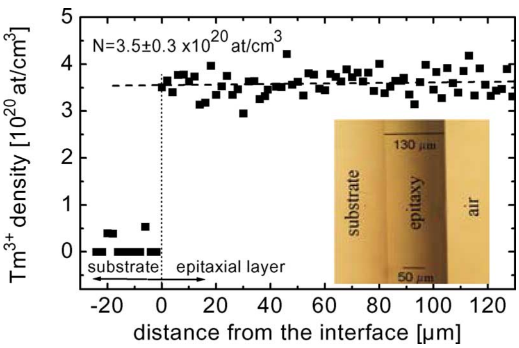

Fig. 3. Tm concentration profile in the Tm:KLuW–KLuW (010) epitaxial composite sample used in the laser. The inset shows an optical microscope image of the layer region.

figure) with transmission T_{OC} =1.5, 3, and 5% were used. M1, M2 and M3 were highly reflecting (HR >99%) from 1800 to 2075 nm and AR-coated on the rear side for high transmission from 780 to 1020 nm. The cavity was designed for longitudinal pumping, normal to the epitaxial layer, with a CW Ti:sapphire laser. The Ti:sapphire laser was tuned by a three-plate intracavity Lyot filter which ensured an output linewidth of <0.2 nm. It delivered a maximum output power of 3.5 W near 800 nm when pumped with 20 W of an all-lines Ar–ion laser. It can be expected that the spectroscopic characteristics of Tm are the same as in bulk KLuW [4]. Thus the pump wavelength was adjusted to 802 nm, the maximum of the absorption for $\boldsymbol{E}//\boldsymbol{N_m}$, and this turned out to be optimum also for the epitaxial Tm-laser.

The epitaxial sample with a total thickness of 1.7 mm (substrate +130-μm-thick doped layer) was mounted in a Cu-block

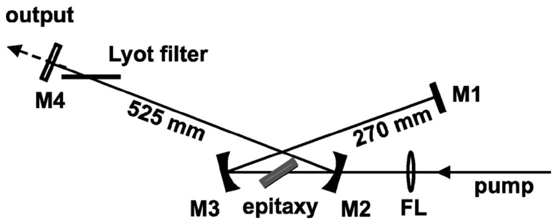

Fig. 4. Cavity setup of the longitudinally pumped epitaxial Tm-laser. FL: AR-coated focusing lens with $f = 70$ mm, M1: plane total reflector, M2-M3: $RC = -100$ mm mirrors, M4: plane output coupler, epitaxy: Tm:KLuW–KLuW sample.

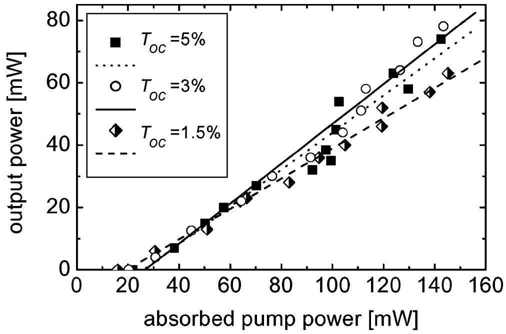

Fig. 5. Input-output characteristics measured for the epitaxial Tm:KLuW laser (symbols) and fits for calculation of the slope efficiencies (lines), for three output couplers.

whose temperature was maintained at 10 °C by circulating water. It was placed between the two folding mirrors under Brewster angle, which determines the laser polarization while the pump polarization was in the same plane. In the position of the epitaxial crystal, the pump spot had a Gaussian waist of 37 μm.

We first studied the laser performance without tuning element in the cavity. The main results are summarized in Fig. 5 and Table I. A maximum output power of 78 mW was obtained for $T_{\mathrm{oc}} = 3\%$. In this case, the pump power incident on the crystal was 1.23 W but the absorbed power measured under lasing conditions was only 144 mW. The lowest threshold was obtained with $T_{\mathrm{oc}} = 1.5\%$ and it corresponded to an incident pump power of 108 mW measured in front of the epitaxial sample. The results, in terms of slope efficiency, are very similar to those obtained with a 5% Tm-doped bulk KLuW [4]. Note, however, that in both cases the slope efficiency is calculated with respect to the absorbed power. The output power obtained with the epitaxial sample is relatively low due to the low absorption under lasing conditions. While the calculated small signal absorption was of the order of 24%, the absorption measured without lasing was strongly bleached and dropped to ≈4%. In the three-level system of Tm, the intracavity intensity modifies the saturation intensity for the pump and the actual absorption increased to about 15% (almost constant with the incident power) but it was still too low. This lead also to the substantially lower thresholds in comparison to bulk Tm:KLuW [4].

The tuning experiments were performed with a birefringent filter (3-mm-thick quartz plate with its optic axis at 60° to the surface). It was inserted in the long cavity arm, close to the output coupler (Fig. 4).

TABLE I
SLOPE EFFICIENCY, LASER WAVELENGTH, AND THRESHOLD OF THE EPITAXIAL Tm-LASER IN DEPENDENCE ON THE T_{OC} USED

T_{OC} [%]	η [%]	λ_L [nm]	threshold [mW]
1.5	48.5	1967	16
3	64	1961	20
5	60	1960	22

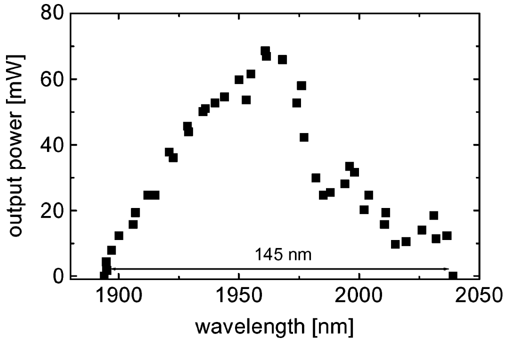

Fig. 6. Tuning of the epitaxial Tm:KLuW laser using an intracavity Lyot filter for $T_{\rm oc} = 3\%$.

Obviously this Lyot filter was not optimized for the present Tm-laser but it still allowed to obtain almost continuous tuning from 1894 up to 2039 nm (Fig. 6). This means a total spectral range of 145 nm with a single output coupler, the FWHM is roughly 60 nm. This result compares very well with the performance of the bulk Tm:KLuW laser [4], although the exact tuning range depends on the doping level, the absorption coefficient, and the output coupler.

IV. CONCLUSION

In conclusion, we demonstrated, for the first time to our knowledge, successful LPE growth and laser operation of Tm-doped composites based on a monoclinic crystal, KLuW. The high optical quality of the sample allowed to obtain slope efficiencies as high as 64% in the CW regime for room temperature laser operation. The epitaxial laser was tunable and its performance was comparable to that of the bulk material. No damage was observed for the incident powers applied in the present work. Future work will aim at higher doping levels and study of the concentration dependence, as well as diode pumping in different cavity geometries.

REFERENCES

[1] A. A. Kaminskii, “Modern developments in the physics of crystalline laser materials,” *Phys. Stat. Sol.*, vol. 200, pp. 215–296, 2003.

[2] S. N. Bagaev, S. M. Vatnik, A. P. Maiorov, A. A. Pavlyuk, and D. V. Plakushchev, “The spectroscopy and lasing of monoclinic Tm:KY$(\mathrm{WO}_4)_2$,” *Quantum Electron.*, vol. 30, pp. 310–314, 2000.

[3] V. Petrov, F. Güell, J. Massons, J. Gavaldà, R. M. Sole, M. Aguiló, F. Díaz, and U. Griebner, “Efficient tunable laser operation of Tm:KGd$(\mathrm{WO}_4)_2$ in the continuous-wave regime at room temperature,” *IEEE J. Quantum Electron.*, vol. 40, no. 9, pp. 1244–1251, Sep. 2004.

[4] X. Mateos, V. Petrov, J. Liu, M. C. Pujol, U. Griebner, M. Aguiló, F. Díaz, M. Galan, and G. Viera, “Efficient 2- μm continuous-wave laser oscillation of Tm^{3+}:KLu$(\mathrm{WO}_4)_2$,” *IEEE J. Quantum Electron.*, vol. 42, no. 10, pp. 1008–1015, Oct. 2006.

[5] A. Giesen, H. Hügel, A. Voss, K. Wittig, U. Brauch, and H. Opower, “Scalable Concept for diode-pumped high-power solid-state lasers,” *Appl. Phys. B*, vol. 58, pp. 363–372, 1994.

[6] N. Berner, A. Diening, E. Heumann, G. Huber, A. Voss, M. Karszewski, and A. Giesen, “Tm:YAG: a comparison between end pumped laser-rods and the ‘thin-disk’-setup,” in *Advanced Solid-State Lasers 1999, OSA Trends in Optics and Photonics*, M. M. Fejer, H. Injeyan, and U. Keller, Eds. Washington, DC: OSA, 1999, vol. 26, pp. 463–467.

[7] U. Griebner, J. Liu, S. Rivier, A. Aznar, R. Grunwald, R. M. Sole, M. Aguiló, F. Díaz, and V. Petrov, “Laser operation of epitaxially grown Yb:KLu$(\mathrm{WO}_4)_2$-KLu$(\mathrm{WO}_4)_2$ composites with monoclinic crystalline structure,” *IEEE J. Quantum Electron.*, vol. 41, no. 3, pp. 408–414, Mar. 2005.

[8] M. C. Pujol, X. Mateos, A. Aznar, X. Solans, S. Suriñach, J. Massons, F. Díaz, and M. Aguiló, “Structural redetermination, thermal expansion and refractive indices of KLu$(\mathrm{WO}_4)_2$,” *J. Appl. Cryst.*, vol. 39, pp. 230–236, 2006.

[9] X. Mateos, R. Sole, J. Gavalda, M. Aguiló, J. Massons, F. Díaz, V. Petrov, and U. Griebner, “Crystal growth, spectroscopic studies and laser operation of Yb^{3+}-doped potassium lutetium tungstate,” *Opt. Mater.*, vol. 28, pp. 519–523, 2006.

[10] M. C. Pujol, M. Aguilo, F. Díaz, and C. Zaldo, “Growth and characterisation of monoclinic $\mathrm{KGd}_{1-x}\mathrm{RE}_x(\mathrm{WO}_4)_2$ single crystals,” *Opt. Mater.*, vol. 13, pp. 33–40, 1999.

[11] A. Aznar, O. Silvestre, M. C. Pujol, R. Sole, M. Aguiló, and F. Díaz, “Liquid-phase epitaxy growth of monoclinic $\mathrm{KLu}_{1-x}\mathrm{Yb}_x(\mathrm{WO}_4)_2/\mathrm{KLu}(\mathrm{WO}_4)_2$ layers,” *Cryst. Growth Des.*, vol. 6, pp. 1555–1558, 2006.