Tunable laser operation of Tm-doped epitaxial layers of monoclinic $KLu(WO_4)_2$ near 2-µm

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Abstract: Epitaxial layers of Tm-doped $KLu(WO_4)_2$ grown on undoped $KLu(WO_4)_2$ substrates are longitudinally pumped by a Ti:sapphire laser at 802 nm and tunable (1894-2039 nm) laser operation is achieved with slope efficiencies as high as 64%.
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1. Introduction

The Tm-doped monoclinic double tungstates, $KRE(WO_4)_2$ (RE=Y, Gd and Lu; hereafter KREW), are distinguished by their largest cross sections, partly due to the strong anisotropy, the higher doping levels possible, and the Stimulated Raman Scattering (SRS) activity. Recently, continuous-wave (CW) laser operation on the $^3F_4 \rightarrow {}^3H_6$ transition was achieved in Tm-doped KYW [1], KGdW [2], and KLuW [3]. With Tm:KLuW, the maximum output power was 4 W and the slope efficiency reached 69% while the tunability extended from 1800 to 1987 nm [3].

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, an effect that is inversely proportional to the thickness of the active element. This 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 [4]. 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 (hereafter 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 [5]. 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. Here we demonstrate efficient laser operation of a Tm:KLuW epilayer and tunability in the 2-µm spectral range, pumping normal to the active layer.

2. Growth of Tm:KLuW/KLuW epitaxial composites

KLuW is a monoclinic crystal with *C2/c* spatial group of symmetry. The unit cell parameters are a=10.576(7) Å, b=10.214(7) Å, c=7.487(2) Å, and β=130.68º(2). Single crystals of KLuW to be used as substrates were grown by the Top Seeded Solution-Growth Slow-Cooling (TSSG-SC) method using $K_2W_2O_7$ as a solvent. More information on the structural and other physical properties as well as on the growth procedure can be found elsewhere [6,7].

The LPE growth was carried out in a special vertical furnace 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 composition of the solution was chosen to be 7 mol % $KLu_{0.95}Tm_{0.05}(WO_4)_2$ – 93 mol % $K_2W_2O_7$, as a compromise, on one hand taking into account our experience with the optical quality of bulk Tm:KLuW crystals [3] and on the other hand aiming at sufficiently high density in order to have highly absorbing layers whose thickness is low enough so that it is otherwise impractical for free-standing active elements. More information on the LPE growth of Tm:KLuW epilayers will be published elsewhere [8].

In analogy with our previous experience with LPE grown Yb:KLuW [9], the best flatness, lowest density of micromorphologies, and 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 polarization $\boldsymbol{E}//\boldsymbol{N_m}$ ensuring maximized interaction cross sections. $\boldsymbol{N_p}$ and $\boldsymbol{N_m}$ denote two of the three orthogonal principal optical axes defined from $n_p<n_m<n_g$. 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.

(a) **(b)**

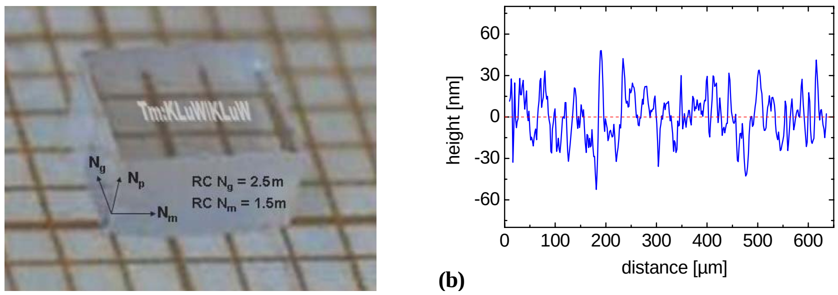

Fig.1: Photograph of the Tm:KLuW/KLuW epitaxial sample used in the laser (a), and roughness profile of its layer surface (b).

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 3.4×3×1.7 mm^3 ($\boldsymbol{N_m}\times\boldsymbol{N_g}\times\boldsymbol{N_p}$). The above composition of the doped layer was derived from Electron Probe Microanalysis with Wavelength Dispersive Spectroscopy (EPMA-WDS). The layer thickness, measured by an optical microscope, was 130 µm. The epitaxial surface was flat over areas sufficiently large for longitudinal pumping, normal to the layer. Figure 1a shows a photograph of the sample and Fig.1b – 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.

3. Laser operation of Tm:KLuW/KLuW

The laser setup used in the present work (Fig.2) was similar to that described in [3]. The astigmatically compensated X-type cavity had a total length of 90 cm. M1, M2 and M3 were highly reflecting (HR > 99.9%) 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 [3]. 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.

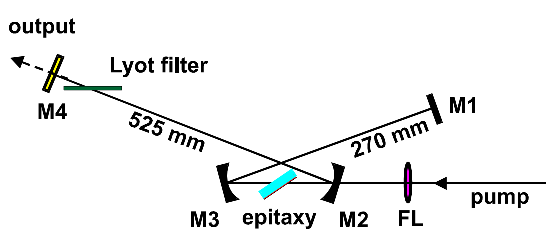

Fig.2: Cavity setup of the longitudinally pumped epitaxial Tm-laser. FL: AR-coated focusing lens with f=70 mm, M1: plane total reflector, M2-M3: RC=-100 mm mirrors, M4: plane output coupler with transmission T_{OC}, epitaxy: Tm:KLuW/KLuW sample.

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

The main results without tuning element in the cavity are shown in Fig.3a. A maximum output power of 78 mW was obtained for T_{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_{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 η, are very similar to those obtained with a 5% Tm-doped bulk KLuW [3]. Note, however, that in both cases η 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 about 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 (16, 20, and 22 mW of absorbed power for T_{OC}=1.5%, 3%, and 5%, respectively), in comparison to bulk Tm:KLuW [3]. The laser wavelength decreased from λ_L=1967 nm (T_{OC}=1.5%) to 1960 nm (T_{OC}=5%), Fig.3a.

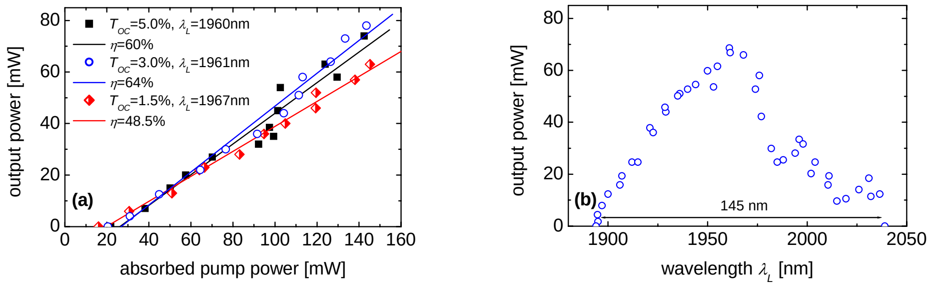

Fig.3: Input-output characteristics measured for the epitaxial Tm:KLuW laser (symbols) and fits for calculation of the slope efficiencies η (lines), for three output couplers (a). Tuning of the epitaxial Tm:KLuW laser using an intracavity Lyot filter, for T_{OC}=3% (b).

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.2). 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.3b. 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 [3], although the exact tuning range depends on the doping level, the absorption coefficient, and the output coupler.

4. Conclusion

In conclusion, we demonstrated, for the first time to our knowledge, successful laser operation of LPE-grown 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 in a spectral range comparable to that obtained with 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.

Acknowledgements

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