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Growth, optical characterization, and laser operation of epitaxial $Yb:KY(WO_4)_2/KY(WO_4)_2$ composites with monoclinic structure

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Epitaxial monoclinic double tungstate laser crystals were grown with high crystalline quality. Based on these Yb-doped composites, laser operation was demonstrated. Continuous-wave laser emission of a Yb:KYW/KYW crystal was achieved at 1030 nm. The 25-μm-thin Yb:KYW layer was pumped at wavelengths near 980 nm by a Ti:sapphire laser. A maximum output power of 40 mW was obtained at room temperature. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1814426]

Yb-doped materials are well suited for building simple and robust diode-pumped lasers in the 1 μm spectral range[1] which have proved their potential in high power operation.[2] The attraction of Yb-doped materials are their small quantum defect and the absence of excited state absorption, upconversion processes, cross relaxation, and concentration quenching. Moreover, the broad absorption bands of these laser materials are covered by high-power InGaAs laser diodes that permit efficient pumping. The small Stokes shift ($\sim 600\ \mathrm{cm}^{-1}$) between absorption and emission reduces the thermal load and increases the laser efficiency.

The magnitudes of the Yb^{3+} absorption and emission cross-section peaks strongly depend on the chosen laser host material. The Yb-doped monoclinic low temperature phases of the double tungstates $KY(WO_4)_2$ (KYW) and $KGd(WO_4)_2$ (KGW) stand out because of their large absorption and emission cross sections.[3] Comparative studies based on the spectroscopic characteristics predicted that they are one of the most promising representatives of this class of materials.[4] KYW can incorporate higher concentrations of Yb^{3+} ions than KGW approaching the stoichiometric composition $KYb(WO_4)_2$ (KYbW)[5] while maintaining the same structure. Yb:KYW, Yb:KGW, and KYbW are strongly anisotropic and all show an absorption maximum near 981 nm which, for light polarization parallel to the N_m-crystallo-optic axis, is about 15 times higher than that of Yb:YAG. In all three of them, efficient laser operation near 1 μm has already been demonstrated.[3,5,6]

The extremely high absorption and emission cross sections of Yb:KYW permit the use of very thin crystals. For such laser crystals even low beam quality pump sources (like most of the laser diodes) can be used, still achieving a good overlap of pump beam and resonator mode. As Yb^{3+} is a quasi-three-level system, poor overlap would not only mean wasted pump power but also cause additional reabsorption of the stimulated emission. Thin disk Yb lasers have already proved their potential for high output powers,[2,6] but have required complicated pump optics to realize several pump passes through the active medium for efficient absorption. This problem can be overcome by using highly doped tungstate materials, but the handling of free-standing crystals is a great technological challenge, in particular for a thickness between 10 and 50 μm. Therefore thin epitaxial structures of highly Yb-doped KYW on KYW substrates seem to be ideal for face-cooled laser concepts including microchip lasers and thin disk laser designs. Our approach is to achieve cw lasing by the use of such epitaxial samples to minimize the reabsorption loss and to pump near the absorption maximum at 981 nm.

Liquid phase epitaxy (LPE) is a well-known technique to obtain homogeneous single crystal layers.[7] It is based on epitaxial growth from solution over a crystalline substrate and allows doping the layers during growth with optically active ions like ytterbium. Moreover it is possible to control the layer thickness by adjusting the temperature and the growth time. Some preliminary results on LPE of KYbW films on KYW substrates were reported only very recently.[8] In the present letter we describe the growth and the properties of an epitaxial Yb:KYW/KYW structure with improved crystal quality at the interface which permitted laser generation to be achieved with a strongly anisotropic (monoclinic) composite structure used as an active medium.

Single crystals of KYW to be used as substrates have been grown by the top seeded solution growth (TSSG) method. The experiments were performed in a vertical cylindrical furnace and the solvent chosen was $K_2W_2O_7$.[9] The solution composition was, in molar ratio, $K_2W_2O_7/KYW = 88/12$. We inserted the reagents, K_2CO_3, Y_2O_3, and WO_3, mixed in the appropriate ratios, weighting about 200 g, in a platinum crucible of an inner diameter of 50 mm. The solution was homogenized by maintaining the temperature at 50 K above the expected temperature of saturation, T_{sat}. The axial thermal gradient in the solution was about 1 K/cm, keeping the bottom hotter than the surface. After homogenization of the solution the T_{sat} has been accurately determined in each run and it is, on average, 920 °C.

Then, the crystal growth process was initiated on seeds in contact with the surface of the solution, which was cooled at a rate of 0.05 K/h by 6 K. Finally, the crystals were removed from the solution and cooled down slowly to room temperature.

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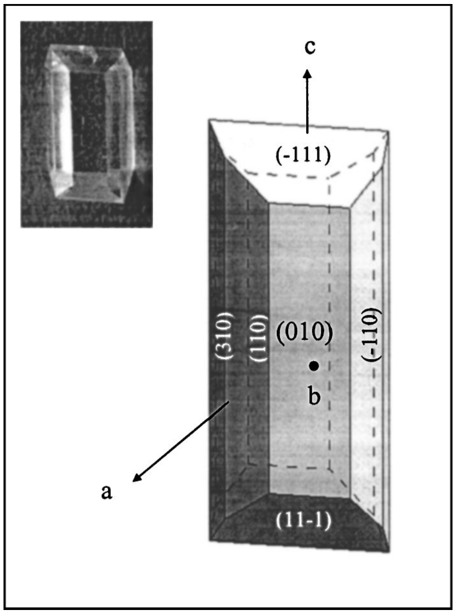

FIG. 1. KYW crystal grown and its morphological scheme.

The KYW substrates obtained were transparent and free from macroscopic defects. The typical dimensions ranged between $3\times3\times6$ and $5\times5\times8$ mm^3 in *a*, *b*, and *c* crystallographic directions, respectively. The unit cell parameters of KYW in the $C2/c$ space group determined at room temperature are,[10] $a=10.6313(4)$ Å, $b=10.3452(6)$ Å, $c=7.5547(3)$ Å, and $\beta=130.752(2)°$ and the unit cell parameters of KYbW amount to $a=10.590(4)$ Å, $b=10.290(6)$ Å, $c=7.478(2)$ Å, and $\beta=130.70(2)°$.[5] It can be seen in Fig. 1 that the KYW morphology is mainly formed by {010}, {110}, {310} and {−111} faces.

The LPE experiments were performed in a vertical furnace with practically no axial gradient to obtain a homogeneous epitaxial layer thickness on every crystal face. The solution was prepared by mixing about 70 g of the reagents in a cylindrical platinum crucible with a 30 mm diameter. The ratio between the solvent, $K_2W_2O_7$, and the solute, $KY_{0.8}Yb_{0.2}(WO_4)_2$, was decreased by 7% as compared to the value in the substrate growth, to perform the epitaxial growth at lower temperatures. This permits a better control of the growth rate according to the solubility curve of KYW in $K_2W_2O_7$.[10]

Once the solution was homogeneous, we accurately determined T_{sat} in each run being, on average, 820 °C, which means 100 °C less than T_{sat} obtained in the substrate growth. Next we studied the kinetics of the growth and dissolution of a seed at different temperatures close to T_{sat}, before the epitaxial growth. This information was very useful for the choice of the temperature and time to perform the epitaxial growth. The substrate was introduced slowly into the furnace and maintained near the surface of the solution for about 30 min to reach a thermal equilibrium with the solution. At the beginning of the epitaxial growth, the temperature of the solution was kept 1 °C higher than T_{sat} with the objective to dissolve the outer layer of the crystal. Thus, the substrate was dipped into the solution at this temperature for 5 min and then the temperature was decreased to 2 °C below T_{sat} for 4 h to grow the epitaxial layer. The crystal rotation velocity during all these procedures (also for TSSG) was kept constant at 40 rpm. Finally we removed the crystal from the solution and took it out slowly from the furnace to avoid cracks due to thermal shocks and the differences of the thermal expansion coefficients between the layer and the substrate. It is important to note that the epitaxial growth takes place on all natural faces of the crystals used as substrates.

TABLE I. Epitaxial film thickness and rate of growth, *R*, for different crystallographic faces measured by SEM.

Face	Thickness (μm)	*R* (μm/h)
(010)	48.1	12
(110)	29.9	7.5
(310)	8.0	2

The thickness of the epitaxial layers was measured by a scanning electron microscope (SEM) using backscattered electrons with the sample cut and polished perpendicular to the *c* crystallographic direction. The results obtained are listed in Table I. The rate of layer growth was faster on the {010} faces followed by the {110} and {310} faces. From the images obtained by SEM a sharp interface between the substrate and the 48 μm thin doped layer, corresponding to a (010) face, can be detected visually (inset Fig. 2).

The substrate and layer composition was determined by electron probe microanalyses with a CAMECA SX-50 equipment. The results obtained show that the Yb content in the layer (11.96×10^{20} Yb^{3+} ions/cm^3) is more or less the same as in the solution, while the Yb content in the substrate is zero, even close to the interface. Thus, the distribution coefficient of Yb^{3+} in these layers is close to unity, which is very favorable for obtaining a homogeneous distribution of ytterbium in the epitaxy. Figure 2 represents the evolution of yttrium and ytterbium concentration at the substrate/layer interface. We observe that there is practically no diffusion of Yb^{3+} into the substrate.

Using an optical microscope we could detect that the surface morphology of these layers was quite good and flat over large areas. There was no essential difference in surface morphology between the different natural faces of the crystal, although the face (010) seems to be the best. The typical defects that appeared were steps of growth in all faces. Furthermore, in {110} and {310} faces some cracks could be identified.

For the laser experiments the (010) faces of the epitaxial crystal were additionally polished with high optical quality. From the measured absorbed pump power we deduced a thickness of the 20% Yb/Y-site KYW layer of 25 μm. The

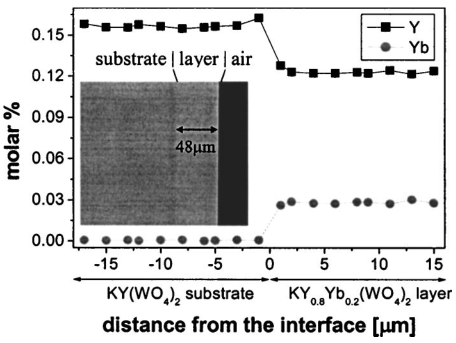

FIG. 2. Evolution of the yttrium (Y) and ytterbium (Yb) concentration at the substrate/layer interface. Inset: SEM micrograph of the epitaxial crystal with the Yb:KYW/KYW interface.

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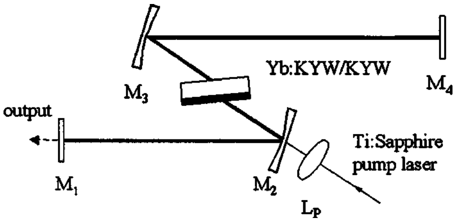

FIG. 3. Laser setup: M$_2$, M$_3$—folding mirrors (RC=100 mm); M$_1$—output coupler; Yb:KYW/KYW—1.6-mm-thick epitaxial crystal; L$_P$—focusing lens.

uncoated 1.6-mm-thick Yb:KYW/KYW epitaxial crystal with an aperture of 3×5 mm^2 was positioned in the laser resonator under Brewster angle to minimize the Fresnel losses. Pump light absorption was maximized by orienting the crystal for the N_m-crystallo-optic axis parallel to the pump light polarization. The experiments were performed with a cw Ti:sapphire laser as the pump source. It was optimized for emission in the range from 960 to 1025 nm with a linewidth <1 nm, delivering more than 2 W output power. We studied a Z-shaped resonator with two folding mirrors in the middle (Fig. 3) to form a 30 μm intracavity beam waist at the position of the epitaxial crystal. No special provision was made for cooling the sample. The Ti:sapphire beam was focused by an $f=62.8$ mm lens through the folding mirror M$_2$ giving an estimated 30 μm pump waist.

Laser operation could be obtained for pump wavelengths between 963 and 997 nm determined by the absorption characteristics of the Yb:KYW layer. Independent of the pump wavelength, the laser emission was centered at 1030 nm (inset Fig. 4). Because of the reduced reabsorption in the thin active layer, the spectral emission corresponds to the maximum of the gain curve. Continuous-wave laser operation was achieved for all pump wavelengths. Approaching the absorption peak near 981 nm, thermal problems occurred and resulted in decreasing the slope efficiency.

The cw laser performance of the Yb:KYW/KYW epitaxial crystal was studied at pump wavelengths λ_p below and above the main absorption peak. Using the setup of Fig. 3, stable laser operation was observed for output coupler transmissions up to 3.5%. Best performance was achieved at $\lambda_p = 984$ nm with a 2% transmission output coupler. The laser threshold of the 25-μm-thin Yb:KYW layer was reached at an absorbed pump power of about 275 mW. The maximum output power amounted to 40 mW resulting in a slope efficiency of 33% with respect to the absorbed pump power (Fig. 4). Applying a chopper with a duty cycle of 4% and comparing the output power for $\lambda_p = 984$ nm and $\lambda_p = 981$ nm, an increase of nearly a factor 4 could be obtained for pumping in the main absorption peak.

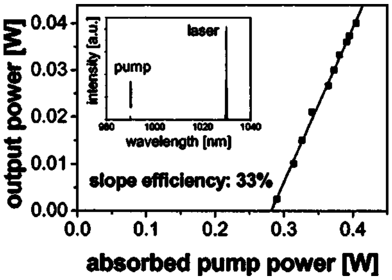

FIG. 4. Output power vs absorbed pump power of the continuous-wave Yb:KYW/KYW laser. Inset: Spectral record of the Yb:KYW/KYW and pump laser emission.

In conclusion, epitaxial monoclinic double tungstate crystals have been grown with high optical quality by the LPE method. A 48 μm thin KYW (010) layer doped with 20% Yb^{3+} as the active ion was grown on a KYW substrate. Laser operation of a Yb:KYW/KYW epitaxial crystal was achieved. Continuous-wave lasing at 1030 nm with an output power of 40 mW was obtained at room temperature. There is great potential for applying such Yb-doped epitaxial KYW crystals in waveguide structures and microchip lasers which profit from the short absorption length.

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