Laser Operation of Epitaxially Grown $Yb:KLu(WO_4)_2$–$KLu(WO_4)_2$ Composites With Monoclinic Crystalline Structure

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***Abstract*—Epitaxial monoclinic double tungstate composites based on the strongly anisotropic $KLu(WO_4)_2$ (KLuW) were grown with high crystalline quality and laser operation of ytterbium was demonstrated for the first time. Highly efficient CW laser emission of an Yb:KLuW–KLuW crystal was achieved near 1030 nm. The 100-μm-thick Yb:KLuW layer was pumped at wavelengths near 980 nm by a tapered diode laser as well as by a Ti:sapphire laser. More than 500 mW of CW output power and slope efficiencies up to 66% were obtained at room temperature without cooling.**

***Index Terms*—Diode pumping, epitaxial layers, monoclinic double tungstates, Yb lasers.**

I. INTRODUCTION

THE growing interest in Yb-doped lasers has been underlined by creating new active materials with Yb^{3+} as a dopant [1]–[3]. Some of these materials have already proved their potential in high-power lasers in the wavelength region near 1 μm. The attractive features of Yb-doped laser crystals include their small quantum defect, the absence of excited state absorption, up-conversion, cross-relaxation and concentration quenching processes, and the broad absorption bands which are covered by high-power InGaAs laser diodes. The relatively small Stokes shift between absorption and emission in the quasi-three-level Yb ion reduces the thermal load and sets an increased quantum limit for the laser efficiency in comparison to the Nd-ion operating in the same wavelength region.

The magnitudes of the Yb^{3+} absorption and emission cross-section peaks strongly depend on the chosen laser host material. Comparative studies based on the spectroscopic characteristics predicted that the Yb-doped monoclinic low-temperature phases of the double tungstates $KY(WO_4)_2$ (KYW) and $KGd(WO_4)_2$ (KGdW) belong to the most promising representatives of this class of materials [4]. They stand out because of their large absorption and emission cross sections and their capability to adopt higher dopant concentrations [5]. $KLu(WO_4)_2$ (KLuW) is isostructural to KYW and KGdW and many relevant properties like refractive index, optical transparency, and thermal conductivity are very similar [6]. The spectral characteristics of Yb:KLuW are also similar to those of Yb:KYW and Yb:KGdW, and the measured fluorescence lifetime of 375 μs for low doped Yb:KLuW is slightly longer than the 300-μs lifetime of Yb:KYW and Yb:KGdW [7]. In fact, we could already demonstrate excellent laser performance of Yb:KLuW in the 1-μm spectral range [7] which was comparable to that reported for Yb:KGdW and Yb:KYW [5]. The highly efficient CW laser operation with this novel Yb-doped monoclinic double tungstate was achieved with 2.2–2.8-mm-thick, 5 at% and 10 at% Yb-doped KLuW samples oriented for polarization $E//N_m$. Output powers of the order of 1 W with pump efficiencies as high as 50% were obtained at room temperature [7].

Due to the closer ionic radii, KYW and KLuW can incorporate higher concentrations of Yb^{3+}-ions than KGdW approaching the stoichiometric composition $KYb(WO_4)_2$ (KYbW) [8] while maintaining the same structure. Yb:KYW, Yb:KGdW, Yb:KLuW, and KYbW are all strongly anisotropic and exhibit an absorption maximum centered near 981 nm. The maximum absorption cross section σ_a for light polarization parallel to the N_m-principal optical axis is about 15 times larger than that of Yb:YAG. For KYbW, the absorption length (1/e level) calculated from σ_a (981 nm) $= 11.7 \times 10^{-20}$ cm^2 is only 13.3 μm. Such exceptionally short absorption lengths permit the use of very thin crystals which considerably reduces the beam quality requirements for the laser diodes used for pumping. To achieve a good overlap of pump beam and resonator mode is especially important in quasithree-level systems like the Yb^{3+} ion because poor overlap would not only mean wasted pump power but will also cause additional reabsorption of the stimulated emission. Thin-disk Yb lasers have already shown their potential for high output powers [2], [9], but up to now require complex pump optics to realize several pump passes through the active medium for efficient absorption. It is this problem that can be overcome by using very thin and highly doped monoclinic double tungstate crystals. Thermo-mechanical limitations, however, set a great technological challenge, in particular for the manufacturing of free standing active elements with a thickness less than 100 μm corresponding to the absorption length. Using epitaxially grown Yb-doped/undoped

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Manuscript received August 9, 2004; revised October 5, 2004. This work was supported by the European Union under Project DT-CRYS, CICyT under Projects MAT2002-04603-C05-03 and FIT-07000-2003-661, and CIRIT under Project 2001SGR00317.

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Digital Object Identifier 10.1109/JQE.2004.842313

TABLE I

Unit Cell Parameters ($C2/c$ Space Group, Room Temperature) of KLuW and KYW in Comparison to the Stoichiometric KYbW as an Ideal Epitaxial Layer. For Comparison the Values for YAG and the Other Known Stoichiometric Yb-Crystal YbAG are Also Included

lattice parameters	*a* [Å]	*b* [Å]	*c* [Å]	*β*	average
KLuW (ref. [17])	10.576(7)	10.214(7)	7.487(2)	130.68(4)°	
KYbW (ref. [18])	10.590(4)	10.290(6)	7.478(2)	130.70(2)°	
Difference	0.13%	0.74%	0.12%	0.02%	0.33%
YAG (ref. [19])	12.0116(3)				
YbAG (ref. [19])	11.9380(5)				
difference	0.61%				0.61%
KYW (ref. [20])	10.6313(4)	10.3452(6)	7.5547(3)	130.752(2)°	
KYbW (ref. [8])	10.590(4)	10.290(6)	7.478(2)	130.70(2)°	
difference	0.39%	0.53%	1.01%	0.04%	0.64%

composites instead of bulk crystals as gain media for thin disk lasers might be prospective, because this technology allows the fabrication of homogeneous epitaxial crystalline layers with perfect structure and suitable composition having a thickness down to the 10-μm range.

Very recently, the first demonstration of a CW thin-disk Yb laser based on an epitaxial crystal as an active material was reported [10]. The authors used a 200-μm-thick Yb:YAG film on YAG as the thin disk. With the 20 at% Yb-doped film and 24 passes for sufficient absorption of the pump light at 940 nm, they achieved 60 W of output power with 30% pump efficiency. Successful fabrication of some thin double tungstate layers with different dopants has also been realized very recently, e.g., by ion implantation (Tm:NaYW) [11] and by laser ablation (Nd:KGdW) [12], but without demonstration of laser operation.

The liquid phase epitaxy (LPE) used in [10] is a well-known technique to obtain homogeneous single crystal layers [13]. The first reports on successful growth of LPE layers were in 1972 [14]. LPE is based on epitaxial growth from solution over a crystalline substrate and allows doping the layers with optically active ions like Yb^{3+} during the growth. 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 [15]. We demonstrated for the first time, to the best of our knowledge, laser operation based on epitaxial double tungstate structures by using a 25-μm 20 at% Yb-doped KYW layer on a KYW substrate crystal [16]. CW lasing at 1030 nm with 40 mW of output power could be achieved.

Essential for laser applications of composite structures is the high optical quality of the epitaxial interface. One important issue is the minimization of the crystal lattice mismatch between the substrate crystal and the epitaxial layer, otherwise stress induced defects can occur. We established that further increase of the Yb-doping concentration in the KYW layer produces a nontolerable stress in the epitaxial structure resulting in cracks at the interface. Consequently we were looking for alternative double tungstate host materials where a reduced stress at the epitaxial interface can be expected. Therefore, KLuW became the focus of our investigations on highly Yb-doped epitaxial double tungstate crystals because of the lower crystal lattice mismatch relative to KYbW in comparison to KYW. The present paper is devoted to characterization and laser investigations of epitaxially grown Yb:KLuW–KLuW composites with optimized crystal quality at the interface. We report highly efficient CW laser operation at room temperature without cooling.

II. Epitaxial Growth

The top-seeded solution growth (TSSG) slow-cooling method was applied to synthesize KLuW single crystals to be used as substrates [17]. The KLuW substrates obtained were transparent and free from macroscopic defects. The typical dimensions ranged between $3 \times 3 \times 6$ and $5 \times 5 \times 8$ mm^3 in a, b and c crystallographic directions, respectively. The unit cell parameters of KLuW and KYW in the $C2/c$ space group determined at room temperature are listed in Table I in comparison to KYbW. Note that for KYbW and KLuW single crystal diffraction measurements are available whereas for YAG, YbAG, and KYW we used powder diffraction data. The closer unit cell parameters of KYbW and KLuW with differences of $0.12 \ldots 0.74\%$ against $0.39 \ldots 1.01\%$ between KYbW and KYW can be seen as a prerequisite for the growth of high-quality epitaxial structures. From Table I, we can also expect a lower induced stress due to the lower lattice mismatch of an epitaxial structure of KLuW and KYbW compared to a composite of YAG and YbAG, the other known Yb-containing stoichiometric crystal with demonstrated laser operation [21].

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. It is important to note that the epitaxial growth takes place on all natural faces of the crystals used as substrates. From Fig. 1 it can be seen that the KLuW morphology is mainly formed by $\{010\}$, $\{110\}$, $\{310\}$ and $\{-111\}$ faces. The thickness of the Yb:KLuW layer, grown on the (010) face, amounted to 130 μm. For the laser experiments, the (010) faces of the epitaxial crystal were additionally polished with high optical quality, resulting in a layer thickness of 100 μm measured by translating the sample in a ZYGO$^{\rm TM}$ interferometer. Using the ZYGO$^{\rm TM}$, we could detect that the surface morphology of the layer of interest was quite good and flat over large areas. Fig. 2(a) shows the surface profile plot of the (010) epitaxial surface. The radius of curvature of the surface

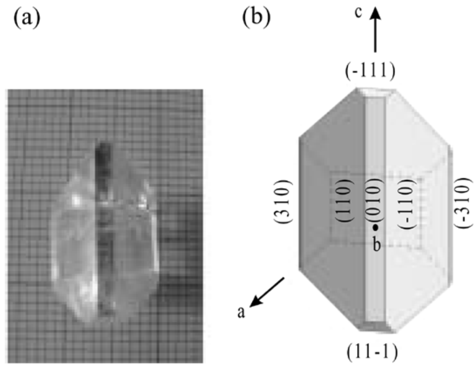

Fig. 1. (a) Photograph and (b) morphological scheme of the KLuW crystal.

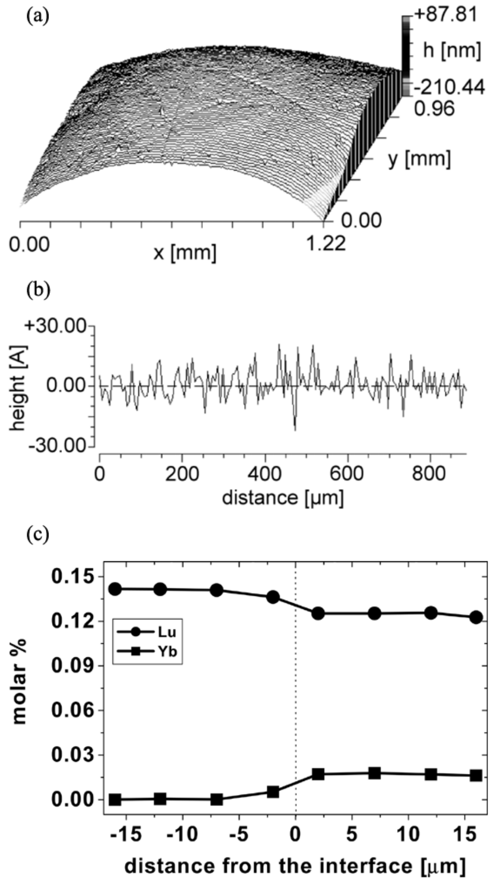

Fig. 2. Characterization of the epitaxial Yb:KLuW–KLuW crystal. (a) Surface profile plot of the (010) epitaxial layer. (b) Roughness of the (010) epitaxial surface. (c) Evolution of the lutetium (Lu) and ytterbium (Yb) concentration at the interface.

is 0.71 m. The surface roughness with an rms value of <0.8 nm presented in Fig. 2(b) emphasizes the excellent optical quality of the (010) epitaxial surface. No cracks at the epitaxial interface could be identified with the ZYGO$^{\text{TM}}$ interferometer.

The substrate and layer composition was determined by electron probe microanalysis with a CAMECA SX-50 equipment. The results obtained show that the Yb content in the layer (7.888×10^{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. From Fig. 2(c), it can be seen that there is practically no diffusion of Yb^{3+} into the substrate.

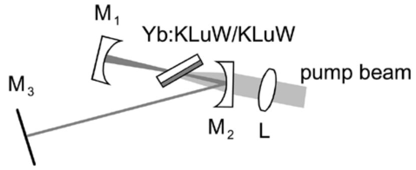

Fig. 3. Experimental setup. M_1: mirror with $ROC = -50$ mm or -100 mm, M_2: mirror with $ROC = -100$ mm; M_3: plane outcoupling mirror; L: $f = 6.28$ cm focusing lens; Yb:KLuW–KLuW—epitaxial crystal.

III. LASER SETUP

The laser experiments with Yb:KLuW–KLuW were performed in a V-type astigmatically compensated resonator as shown in Fig. 3. The 1.2-mm-thick epitaxial sample was positioned in the focal region between the two focusing mirrors which formed a 30-μm intracavity beam waist. It was uncoated and inclined under Brewster angle to minimize the Fresnel losses. The Yb:KLuW–KLuW crystal was oriented for propagation along the $\boldsymbol{b}(N_p)$ axis with faces parallel to the $N_m - N_g$ plane and polarization along the N_m principal optical axis. Deviations from this orientation occurred only as a consequence of the Brewster geometry used but did not essentially modify the absorption and gain spectral profiles. The polarization choice plays a very important role for the optimization of the net amplification. The folding mirror M_2 (radius of curvature $ROC = -100$ mm) transmitted $\approx 99\%$ of the pump radiation near 980 nm and was highly reflective for the laser radiation. The end mirror M_1 with a $ROC = -50$ mm was highly reflective only for the laser wavelength, alternatively a mirror with $ROC = -100$ mm which was highly reflecting also for the pump radiation was applied in order to study double-pass pumping. The cavity length was about 67 and 72 cm, respectively. In both cases, the calculated mode size in the center of the stability range was about 40 μm for the Gaussian waist. The plane output coupler M_3 had a transmission between 1% and 10% near 1030 nm. No special care was taken for good thermal contact or cooling of the sample.

We used a diode laser or a Ti:sapphire laser as pumping sources but only in the latter case was it possible to realize a double pass pumping utilizing the residual radiation which was not absorbed in the first pass. The tunable Ti:sapphire laser was optimized for emission in the range from 960 to 1025 nm with a linewidth < 1 nm, delivering more than 2.5 W of output power. For the diode-pumped operation, a tapered diode laser (TDL) was used [22], delivering up to 2 W at an $M^2 < 3$ for the slow axis emission. Temperature tuning of this laser was possible between 975–982 nm. The emission of the TDL with a spectral bandwidth of only 1 nm was stabilized near 980 nm by feedback of a small amount of the radiation $(< 0.02\%)$ using a reflection grating. Due to the excellent beam quality of the TDL, relatively simple beam shaping optics were required, and a good overlap of pump and cavity modes could be realized. The astigmatic emission of the TDL was formed by an aspherical ($f = 4.5$ mm) and a cylindrical ($f = 40$ mm) lens

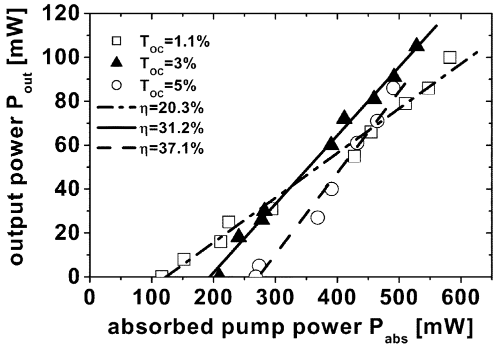

Fig. 4. Output power $P_{\rm out}$ versus absorbed pump power $P_{\rm abs}$ (symbols) of the TDL-pumped CW Yb:KLuW–KLuW laser obtained at the optimum $\lambda_P = 981.3$ nm, and linear fits (lines) for calculation of the slope efficiency η for three different output couplers $(T_{\rm OC})$. In all cases $\lambda_L \approx 1030$ nm.

to achieve a nearly collimated pump beam. The pump beam was focused in both cases by an $f = 62.8$ mm AR-coated lens (Fig. 3) through the folding mirror M_2 onto the crystal giving an estimated 30-μm pump waist in the case of Ti:sapphire pumping and slightly larger for the TDL pump source.

The measured single pass low-signal absorption of the 10% Yb–Lu-site KLuW layer at 981.5 nm amounted to 64% in good agreement with the calculated value of 64.7% for a 100 μm thickness under Brewster angle ($\sigma_a = 11.8 \times 10^{-20}$ cm^2 at 981.5 nm [7]).

IV. RESULTS AND DISCUSSION

In the laser experiments with TDL pumping, the end mirror M_1 (Fig. 3, $ROC = -50$ mm) was highly transmittive for the pump radiation and the absorbed power in the single pass could be precisely measured. CW laser operation could be obtained for output coupler transmissions $(T_{\rm OC})$ between 1.1% and 10%. The output power $(P_{\rm out})$ versus the absorbed pump power $(P_{\rm abs})$ for three different $T_{\rm OC}$ is shown in Fig. 4. The laser threshold achieved with the 100-μm-thin Yb:KLuW layer was as low as $P_{\rm abs} \approx 120$ mW for the 1.1% output coupler. Note that the thresholds we measured with different outcouplers were three to five times lower than with the bulk Yb:KLuW samples [7] where reabsorption forced the laser to oscillate at longer wavelength λ_L. At the maximum applied pump power (1.25 W incident on the sample), the maximum output power amounted to 105 mW and the pump efficiency with respect to the absorbed power reached 20% $(T_{\rm OC} = 3\%)$. The highest slope efficiency with respect to the absorbed power, $\eta = 37.1\%$, was achieved with the $T_{\rm OC} = 5\%$ output coupler.

The actual absorption depends on the depletion effect and can be substantially lower than the small-signal value. It depends, however, also on the output coupler transmission since the different intracavity power produces a different recycling effect which counteracts the depletion [23]. Fig. 5(a) shows the absorption dependence on the incident pump power $P_{\rm inc}$ for three output couplers and also without lasing. The incident pump intensity is comparable to or exceeding the saturation intensity and since the active layer thickness is smaller than the absorption length the absorption depletion effect can be clearly observed [Fig. 5(a)]. Note that this was not the case when pumping thick bulk samples of Yb:KLuW with the same TDL [7]. It can be seen from Fig. 5(a) that the depletion effect is strongest when lasing is interrupted (in the $M_2{-}M_3$ arm in Fig. 3). Lower T_{OC} corresponds to increased intracavity power $P_{\rm intr}$ and consequently the absorption increases [Fig. 5(b)]. But for a given T_{OC} the dependence on $P_{\rm inc}$ is not strong because $P_{\rm intr}$ also increases with $P_{\rm inc}$ [Fig. 5(a) and (b)].

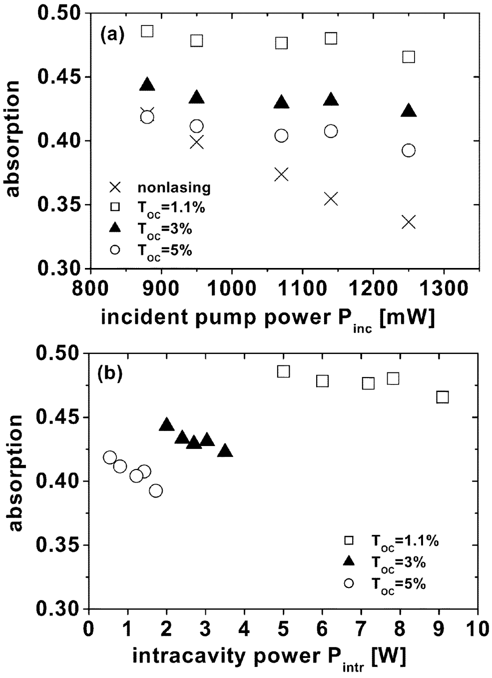

Fig. 5. Estimated single-pass absorption in the case of TDL pumping versus (a) the incident pump power $P_{\rm inc}$ on the epitaxial crystal and (b) the intracavity laser power $P_{\rm intr}$.

With the Ti:sapphire laser used as a pump source in the same single-pump-pass cavity configuration, a much better performance was achieved which was due to its nearly diffraction-limited beam quality and the improved mode matching between the pump and laser modes as compared to the TDL pumping. The CW output characteristics are presented in Fig. 6. We limited the incident pump power applied to 1.85 W. The maximum measured output power of 415 mW corresponds to a maximum efficiency of 55% with respect to the absorbed pump power $(T_{\rm OC} = 3\%)$. The slope efficiency with respect to the absorbed power increases with $T_{\rm OC}$ reaching a maximum value of $\eta = 66\%$ $(T_{\rm OC} = 10\%$, Fig. 6). Both the pump and slope efficiencies exceed those we recently reported for a 2.2-mm-thick 10 at% Yb-doped bulk KLuW in a similar pump and laser configuration [7]. This is attributed to the strongly reduced reabsorption which leads to about 4 times lower thresholds in the case of the epitaxial sample (about 70 mW in Fig. 6) and to shorter laser wavelengths λ_L. Note that the dependence of λ_L on $T_{\rm OC}$ in Fig. 6 can be explained by stronger absorption depletion and more homogeneous pumping along the beam path when increasing $T_{\rm OC}$.

The dependence of the actual absorption on the intracavity intensity is more pronounced in the case of Ti:sapphire laser pumping because of the stronger pump induced absorption depletion (Fig. 7). The stronger saturation due to the smaller pump spot size can be seen by comparison of the absorption with

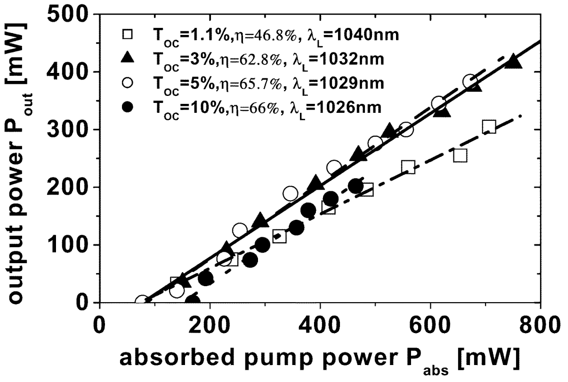

Fig. 6. Output power P_{out} versus absorbed pump power P_{abs} (symbols) of the Ti:sapphire laser pumped CW Yb:KLuW–KLuW laser obtained at the optimum $\lambda_P = 981.5$ nm, and linear fits (lines) for calculation of the slope efficiency η for four different output couplers T_{OC}.

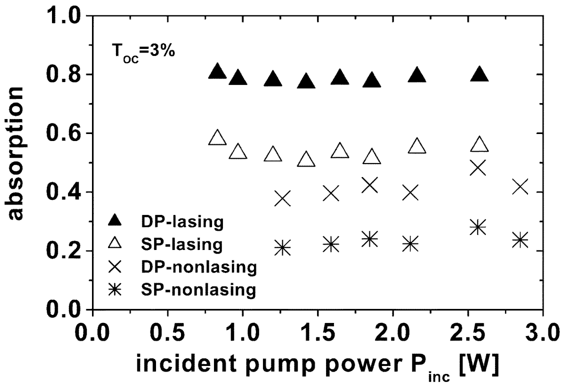

Fig. 8. Estimated single-pass (SP) and double-pass (DP) absorption with lasing and without lasing in the case of Ti:sapphire laser pumping with feedback versus the calculated incident pump power P_{inc} on the epitaxial crystal.

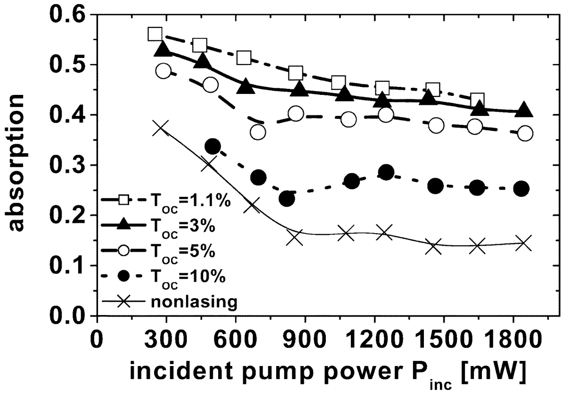

Fig. 7. Estimated single-pass absorption in the case of Ti:sapphire laser pumping versus the incident pump power P_{inc} on the epitaxial crystal.

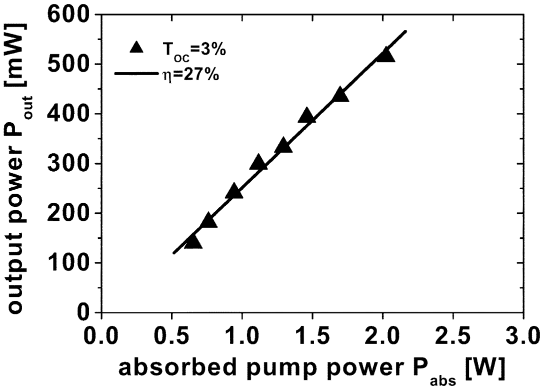

Fig. 9. Output power P_{out} in the case of Ti:sapphire laser pumping with feedback versus the calculated absorbed pump power in double pass P_{abs}.

lasing interrupted in the cases of diode and Ti:sapphire laser pumping. In the case of Ti:sapphire laser pumping, the absorption is depleted at relatively low incident powers and the further dependence on P_{inc} is saturated.

We examined the influence of thermal effects by employing a chopper with a 1:10 duty cycle. We observed only a weak effect of about 10% at the maximum applied pump powers (see Fig. 6), i.e., the maximum average output power achieved with the chopper was 45 mW.

The Ti:sapphire laser beam quality and the close pump and laser wavelengths allow in principle simple double pass pumping by using a retroreflector for the pump radiation. In general the second pass can help to suppress the reabsorption effect. In the present case, we were motivated to try this on one hand by the reduced single-pass absorption in comparison to the small-signal case (Fig. 7) and on the other hand by the observation that feedback effects caused by mirror M_1 (Fig. 3) could increase the pump power leading to coupled cavities or equivalently the Yb:KLuW–KLuW laser could be considered as being partially intracavity pumped. To investigate this possibility we employed another mirror M_1 with $ROC = -100$ mm which was highly reflective both for the pump and laser radiation. The feedback effect on the Ti:sapphire pump laser strongly depended on its output power and decreased at higher powers.

In the CW regime, we applied a maximum pump power of 950 mW measured at the output of the Ti:sapphire laser. The actual incident pump power was calculated by monitoring a weak retroreflection from the lens (Fig. 3). As can be seen from Fig. 8, the absorption without lasing was strongly saturated but increased with the second pass. Lasing not only increased the actual absorption but as a consequence of this decreased the feedback and the maximum incident pump power was lower ($P_{\mathrm{inc}} = 2.85$ W without lasing and $P_{\mathrm{inc}} = 2.58$ W with lasing, respectively).

The maximum output power P_{out} obtained from the Yb:KLuW–KLuW laser with pump feedback reached 515 mW with $T_{\mathrm{OC}} = 3\% (\lambda_L = 1030\ \mathrm{nm})$ (see Fig. 9). This corresponds to a maximum pump efficiency of 25.5%. The maximum P_{out} slightly dropped with $T_{\mathrm{OC}} = 5\%$ to 480 mW and amounted to 400 mW with the 1.1% output coupler.

Using the same chopper with 1:10 duty cycle, we increased then the Ti:sapphire laser pump power measured directly at its output to 2 W. The feedback increased this value to $P_{\mathrm{inc}} = 3$ W. With the 3% output coupler, the average output power of the epitaxial laser was 100 mW and the wavelength remained unchanged, $\lambda_L = 1030$ nm. The estimated double-pass absorption leads in this case to a pump efficiency of 40% which means a substantial increase in comparison with Fig. 9. Notwithstanding the fact that the results in terms of efficiency are worse in comparison to the case of pumping without feedback (presumably because of poor mode matching and overestimation of the absorption in the second pass), the feedback provides higher output powers. While this type of quasi-intracavity pumping is of practical interest because it increases the overall (relative to the Ar laser) pump efficiency about two times, the feedback mechanism is expected to play a minor role at increased dopant concentrations. From the maximum output power obtained with the chopper, it can be concluded that CW output powers in excess of 1 W can be expected from this epitaxial laser once a proper cooling is provided.

It is interesting to note that, even without cooling, no damage of the epitaxial crystal occurred regardless of the high power levels (intracavity intensity exceeding 1 MW/cm^2 with the $T_{OC} = 1.1\%$ outcoupler).

In conclusion, we studied for the first time, to the best of our knowledge, CW lasing in the 1-μm range of Yb^{3+}-doped epitaxial layers using the monoclinic host KLuW as a substrate. Pump efficiencies as high as 55% and CW output powers exceeding 500 mW were achieved without active cooling the crystal. This is an improvement of about one order of magnitude in comparison to our initial results obtained with Yb:KYW–KYW epitaxies [16]. In the present work we also demonstrated diode pumping of the epitaxial laser near 980 nm. Work is in progress to increase the doping level of the epitaxial layer toward the stoichiometric composition KYbW.

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