Continuous-Wave Laser Oscillation of Yb^{3+} in Monoclinic $KLu(WO_4)_2$

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***Abstract*—The strongly anisotropic monoclinic double tungstate crystal $KLu(WO_4)_2$ was doped with Yb^{3+} ions and shown to be a highly efficient active laser medium in the 1-μm spectral range for continuous-wave room-temperature operation, very suitable for pumping with InGaAs laser diodes.**

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

I. Introduction

The crystal structure of the monoclinic low-temperature phase of $KLu(WO_4)_2$ (KLuW) was studied as early as 1968 [1]. The unit cell parameters determined in the $C2/c = C_{2h}^6$ space group by powder diffraction measurements amount to [2]: $a = 10.592(3)$ Å, $b = 10.236(6)$ Å, $c = 7.498(1)$ Å, and $\beta = 130.75(2)°$. The density of KLuW is 7.80 gcm^3 and the melting point is 1090 °C [1]. Polarized infrared and Raman spectra of KLuW were analyzed in [3]. Efficient high-order Stokes and anti-Stokes stimulated Raman scattering (SRS) in the visible and near-infrared was observed for the two SRS-active modes at 907 and 757 cm^{-1} [4]. Other properties of KLuW like the thermal conductivity ($\approx 3\,Wm^{-1}K^{-1}$), hardness, optical transparency, and refractive index are very similar to those of the isostructural $KY(WO_4)_2$ (KYW) [4]. Laser emission of Er:KLuW was demonstrated for the 0.85-, 1.74- and 2.81-μm transitions [5], the luminescence properties and laser operation of Ho:KLuW for the 2.08- and 2.94-μm lines were studied in [6]–[8], but most of the spectroscopic and laser studies were devoted to Nd:KLuW [8]–[11], where the 1.07- and 1.35-μm lines were investigated and continuous-wave (CW) generation at 1070.2 nm achieved with diode pumping [11].

While in the case of Nd-doping no specific advantages of KLuW over the analogous passive hosts KYW and $KGd(WO_4)_2$ (KGdW) could be revealed [10], [11], the situation with Yb-doping is substantially different. KYW can be doped with very high concentrations of Yb reaching the stoichiometric structure $KYb(WO_4)_2$ (KYbW) [12] with practically no concentration quenching. Such highly Yb-doped materials are potentially interesting for thin film laser designs which profit from the relaxed requirements to the pump laser beam quality and the possibility for efficient transversal cooling, especially in the high-power regime, thanks to the small quantum defect and the absence of excited state absorption and up-conversion processes characteristic of the Yb ion. Monoclinic double tungstates possess the additional advantage of exceptionally large absorption and emission cross sections, however, their poor thermomechanical properties set a challenge for the manufacturing of active elements with a thickness less than 100 μm corresponding to the absorption length. Epitaxial growth of doped/undoped composites is a promising solution to the problem, however, the crystal lattice mismatch of KYW and KYbW seems to be the basic limitation on the achievable interface quality [13]. The present study of KLuW as a new host for the Yb^{3+} ion was motivated on the first place by the closer unit cell parameters of KYbW and KLuW (0.02%…0.5% differences against 0.4%…1% between KYbW and KYW [13]) which is seen as a prerequisite for

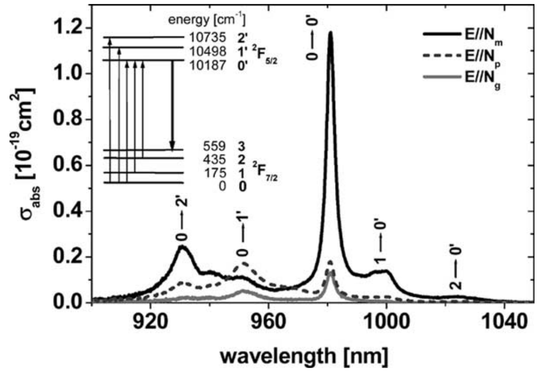

Fig. 1. Absorption cross section of Yb^{3+} in KLuW for the three polarizations and energy level scheme (inset) with the absorption and lasing transitions.

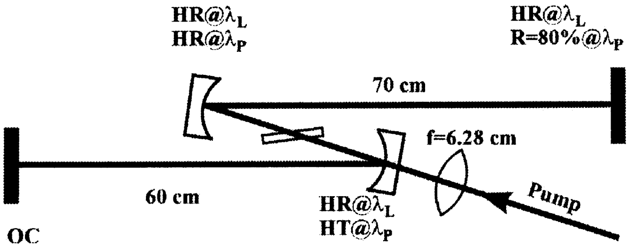

Fig. 2. Laser setup (OC: output coupler).

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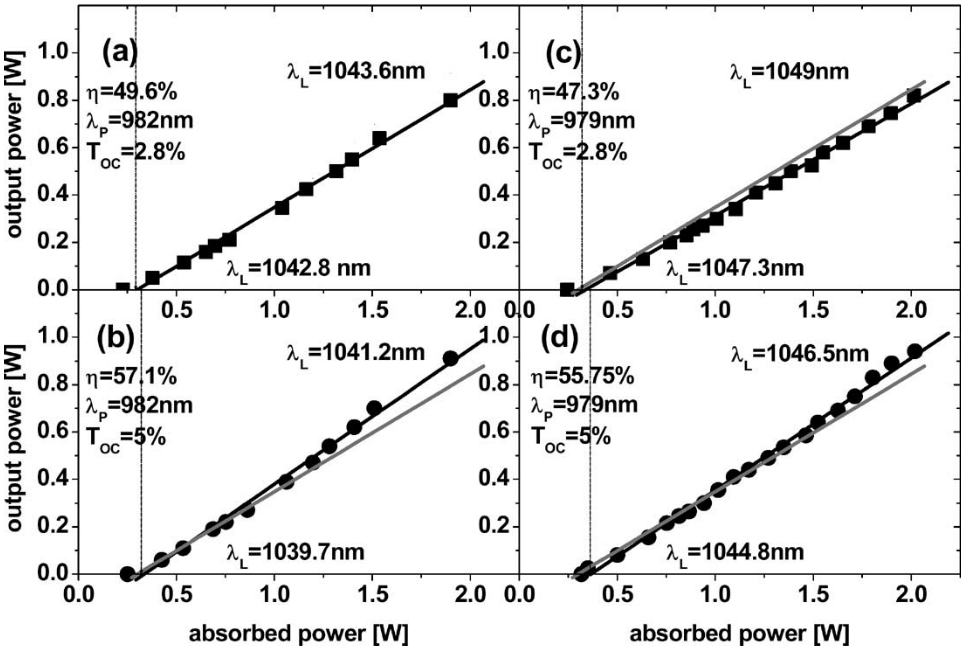

Fig. 3. Output power P_{out} versus absorbed power P_{abs} in double pass (symbols) obtained at the corresponding optimum λ_P, and linear fits (black lines) for calculation of the slope efficiency η for (a)–(b) the 5% and (c)–(d) 10% Yb-doped KLuW samples and two different output couplings (T_{OC}). The wavelength λ_L is indicated both at threshold (bottom values) and at maximum P_{out} (top values). The gray lines in (b), (c), and (d) are only guiding and have the same slope as the black line in (a). The thin vertical lines indicate the extrapolated thresholds.

the growth of high-quality epitaxial structures. Comparison of the thermomechanical properties (expansion, conductivity) of highly Yb-doped KLuW and KYW is planned for the near future.

II. EXPERIMENTAL

We grew undoped and Yb-doped single crystals of KLuW by the top-seeded solution growth (TSSG) slow-cooling method as previously described for growing the isostructural KGdW [14]. The crystal growth conditions were similar to those for KGdW, KYW, and KYbW: The solvent used was $K_2W_2O_7$ and the seed orientation was the b crystallographic axis [15]. The most remarkable difference concerned the morphology, KLuW crystals did not develop the (010) face like the other monoclinic double tungstates. The weight of the crystals was between 3 and 4.5 g, and the dimensions were 10…12 mm, 5…9 mm, and about 13 mm along the a^*, b, and c crystallographic axes, respectively. The single crystals grown were free of inclusions and macrodefects.

The absorption cross section of Yb^{3+} in KLuW was derived from room-temperature optical density measurements with 1 nm instrumental resolution (Fig. 1). The three polarizations correspond to the three orthogonal principal optical axes N_g, N_m and $N_p//b$ defined from the relation $\mathrm{n}_g > \mathrm{n}_m > \mathrm{n}_p$ for the refractive indices. N_g is located at $\approx 18.5^\circ$ from the crystallographic axis c in the clockwise direction when the b-axis is pointing toward the observer. The main absorption line with a maximum cross section of $\sigma_{\mathrm{abs}} = 1.18 \times 10^{-19}$ cm^2 for polarization $E//N_m$ is centered at 981.1 nm and has a FWHM of 3.6 nm. The emission cross section at this wavelength calculated by the reciprocity method amounts to 1.47×10^{-19} cm^2. The fluorescence lifetime measured with a low doped (0.5 mol% Yb) KLuW to minimize radiation trapping amounted to 375 μs which is slightly larger than for KYW (300 μs).

For laser generation, we tested Yb:KLuW samples with 5 and 10 mol% Yb-content in the solution. The concentration of Yb ions was determined by electron-probe microanalysis (EPMA) with Cameca SX 50 equipment. The measured values of 4.300×10^{20} cm^{-3} and 8.258×10^{20} cm^{-3} for the 5% and 10% Yb-doping correspond to compositions of $\mathrm{KLu}_{0.932}\mathrm{Yb}_{0.068}(\mathrm{WO}_4)_2$ and $\mathrm{KLu}_{0.8700}\mathrm{Yb}_{0.1300}(\mathrm{WO}_4)_2$, respectively. A sample of Yb:KYW with 10 mol% Yb in the solution [8.172×10^{20} cm^{-3} Yb ion density in the crystal or a composition of $\mathrm{KY}_{0.8714}\mathrm{Yb}_{0.1286}(\mathrm{WO}_4)_2$] was available as a reference. The thickness of the 5% and 10% Yb:KLuW and the 10% Yb:KYW samples was 2.8, 2.2, and 2.37 mm, respectively. All samples were cut and polished with their parallel faces normal to the N_p-principal optic axis. By rotation around this axis it was possible to align them for polarization parallel to the N_m-principal optic axis in which case maximum absorption (Fig. 1) and gain are expected.

The samples were inserted under Brewster angle between the folding mirrors (*RC* = –10 cm) of a standard, astigmatically compensated *Z*-shaped resonator (Fig. 2). The pump radiation was focussed by an AR-coated lens through one of the folding mirrors which was highly transmitting near 980 nm while the second folding mirror was highly reflecting both at the laser (λ_L) and pump (λ_P) wavelengths. The latter permitted to pump the active medium from the back side in a second pass by 80% retroreflection at the total plane reflector terminating this arm (Fig. 2) of the pump radiation nonabsorbed in the first pass. No special care was taken to cool the crystal. In the other arm of the cavity a plane output coupler (OC) with transmission T_{OC} =1.5%…10% was used. We applied two pump sources:

TABLE I
MAXIMUM OUTPUT POWER P_{OUT} AND PUMP EFFICIENCY η_0
ACHIEVED WITH YB:KLUW

	5% Yb:KLuW P_{abs}=1.90 W, λ_P=982 nm		10% Yb:KLuW P_{abs}=2.02 W, λ_P=979 nm	
T_{OC} [%]	P_{out} [mW] / η_0 [%]	λ_L [nm]	P_{out} [mW]/η_0 [%]	λ_L [nm]
1.5	465 / 24.5	1047.2	490 / 24.3	1051.3
2.8	800 / 42.1	1043.6	820 / 40.6	1049.0
5	910 / 47.9	1041.2	940 / 46.5	1046.5
10	965 / 50.8	1033.3	1010 / 50.0	1041.0

the first was a home-made broadly tunable CW Ti:sapphire laser delivering more than 2.5 W near 980 nm when pumped by 25 W (all lines) of an Ar ion laser. This laser was tuned with an intracavity birefringent filter reducing the linewidth to less than 1 nm. The second pump source was a tapered InGaAs diode laser delivering up to 2 W of output power at 978 nm and tunable between 975 and 981 nm by the temperature, with an M^2 factor for the slow axis emission of less than 3 ($1/e^2$-value). More details on this laser diode can be found elsewhere [16].

III. RESULTS AND DISCUSSION

With $T_{\mathrm{OC}} = 2.8\%$ the Yb:KLuW laser threshold could be reached for 965 nm $< \lambda_P <$ 1005 nm. The optimum Ti:sapphire laser pump wavelength for the 5% Yb-doped KLuW sample was $\lambda_P = 982$ nm and for the 10% Yb-doped KLuW sample it was $\lambda_P = 979$ nm. On the basis of the absorption spectrum in Fig. 1 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. In both samples, for a pump beam waist of the order of 30 μm we observed saturation of the absorption at λ_P: at maximum pump powers, 450–480 mW and 520–550 mW were transmitted behind the 5%-doped and the 10%-doped KLuW samples, respectively, while near the threshold the whole pump power was absorbed in the first pass. The results obtained at the optimum λ_P with the two Yb:KLuW samples are shown in Fig. 3 for two values of T_{OC}: 2.8% (a, c) and 5% (b, d). The maximum output powers P_{out} and pump efficiency with respect to the absorbed power η_0 are summarized in Table I. The four output couplers used were designed for 1064 nm and the specified T_{OC} is for an average wavelength of 1040 nm, i.e., the actual T_{OC} is slightly higher(lower) for λ_L below(above) 1040 nm. 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 Fig. 3 and Table I, having in mind the slightly different wavelengths and absorbed powers, are almost identical in terms of conversion efficiency and threshold. Moreover, the comparison with the 10% Yb-doped KYW reference sample, whose thickness corresponded to an optical density equal to that of the 10% Yb-doped KLuW sample, indicated the same optimum λ_P and output characteristics as in Table I.

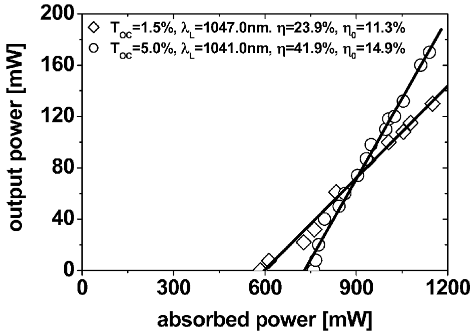

Fig. 4. Output power P_{out} versus absorbed power P_{abs} (symbols) obtained at the optimum $\lambda_P = 980$ nm with laser diode pumping, and linear fits (lines) for calculation of the slope efficiency η for the 5% Yb-doped KLuW sample and two different output couplings (T_{OC}). The wavelength λ_L is indicated at maximum P_{out}.

Pumping with the laser diode was applied only to the 5% Yb-doped KLuW sample because of its lower absorption. At the optimum pump wavelength of $\lambda_P = 980$ nm the recorded output power versus absorbed power is shown in Fig. 4. No saturation of the absorption took place in this case because of the lower pump intensity and the total pump power incident on the sample was absorbed in the first pass. This resulted in increased reabsorption losses and therefore in lower slope and maximum efficiencies, and higher oscillation thresholds. The almost unchanged λ_L in Fig. 4 in comparison to Fig. 3 and Table I indicate, however, that the laser performance is affected to a greater extent by the imperfect overlap of the pump and laser waists in the crystal.

We studied, for the first time to our knowledge CW lasing in the 1-μm range of Yb^{3+} in the monoclinic host KLuW achieving conversion efficiencies as high as 50% and output powers of the order of 1 W without active cooling the crystal. Diode pumping at 980 nm was also demonstrated.

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