Spectroscopy of Tb:glass

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Abstract

Absorption and emission spectra are measured in Tb-doped Duran glass samples. The absorption cross-section of the $^7F_6 \rightarrow {}^5D_4$ pump transition is determined to be $\sigma_{max} = 1.23 \cdot 10^{-22}$ cm^2. The intensity of the green fluorescence from $^5D_4 \rightarrow {}^7F_5$ is measured as a function of 488 nm pump intensity. Within the range of intensity used in the experiment no deviation from linearity can be detected. Also in the intensity of UV-lines measured as a function of pump intensity no quadratic characteristic can be detected even if they originate from up-conversion pumping.

Introduction

The first terbium laser was found by Bjorklund in 1967 [1], where terbium was in liquid form of a 2.5 mMol solution of terbium-chelate. The emission occurred in the transition $^5D_4 \rightarrow ^7F_5$ at a wavelength of 547 nm and the solution was pumped with a flash lamp [2]. Other hosts that allow laser emission at 544 nm are Tb (10 at.%):Gd (10 at.%): yttrium lithium fluoride (YLF) [3, 4] and sodium terbium borate [5]. There is also a Tb:YLF crystal with laser activity in the transition of $^7F_3 \rightarrow ^7F_5$ at 4.1 μm, pumped with a Ho:YLF laser at 2.06 μm [6].

As yet, however, no laser activity has been found in Tb:glass. Glass fibres with a small core diameter allow maintenance of high pump intensity over very long distance. This is a promising prerequisite for laser action. Also the lifetime of $^5D_4$ of 2.01 ms [7] is a good condition for laser action. Nevertheless, all attempts to reach laser threshold by pumping the $^7F_6 \rightarrow ^5D_4$ transition at 488 nm have been without success as yet. Possible reasons are a small absorption cross-section at 488 nm, a small emission cross-section in the $^5D_4 \rightarrow ^7F_5$ transition or re-absorption of the emission line in an excited state. Further, the quality of the used sol-gel based fibres in [7, 8] was possibly not high enough and led to scattering losses. It is therefore interesting to study the spectroscopic properties in glass samples of high quality.

In this report we describe absorption measurements and excitation of fluorescence by 488 nm pumping in Tb:glass. The emission is studied primarily in the green $^5D_4 \rightarrow ^7F_5$ transition in the range of λ = 542 nm, but also in the near UV transitions of $^5D_3 \rightarrow ^7F_J$.

Experiments and results

The investigated samples are similar to the commercially available DURAN glass of Schott. They are, however, made from high-purity materials as required for fibre production.

Glass composition	82 mol% SiO_2-13 mol% B_2O_3-1 mol%Al_2O_3-4 mol% (Na_2O/K_2O)
Index of refraction n_e	1.473 @ 546.1 nm
Abbe number ν	66
Density	2.22 g/cm^3
Thermal expansion	2.2 ppm/°K
Transformation temperature	530°C
Doping	$1{\cdot}10^{20}$ Tb^{3+}/cm^3; $1{\cdot}10^{19}$ Tb^{3+}/cm^3; $1{\cdot}10^{18}$ Tb^{3+}/cm^3

Tab. 1: Properties of the investigated samples

Absorption spectroscopy

The transmittance of the Tb:glass samples with 1 cm thickness was investigated in a spectrophotometer (Perkin Elmer Lambda 19). In the Tb:glass cube with the strongest dopant concentration ($1{\cdot}10^{20}$ Tb^{3+}/cm^3) two absorption lines in the IR range at 1940 nm ($^7F_6 \rightarrow {}^7F_1$) and 1885 nm ($^7F_6 \rightarrow {}^7F_0$) respectively, can be assigned to Tb^{3+}. Other absorption lines at 1.4 µm, and above 2 µm can be assigned to OH (cf. Fig. 1).

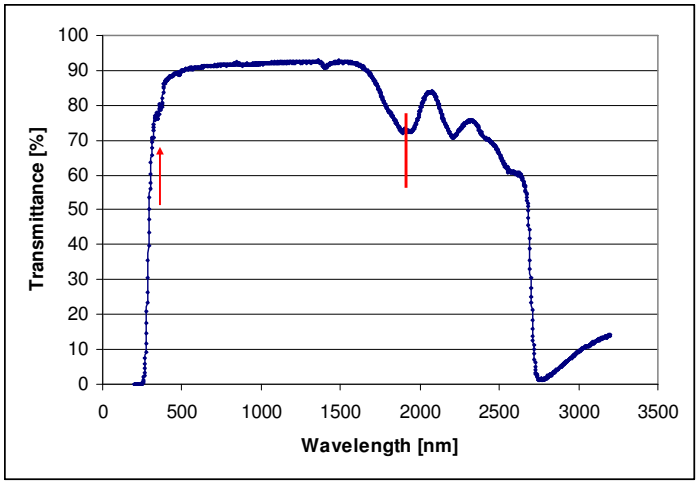

Fig. 1: Transmittance of glass doped with a Tb^{3+} concentration of $1{\cdot}\ 10^{20}$ cm^{-3} and a thickness of 1 cm in the range of 300 nm to 3200 nm. The arrow shows the near UV region displayed in Fig. 3.

In the visible part of the spectrum the blue-green absorption line is found at 485nm ($^7F_6 \rightarrow {}^5D_4$). A more detailed spectrum is shown in Fig. 2.

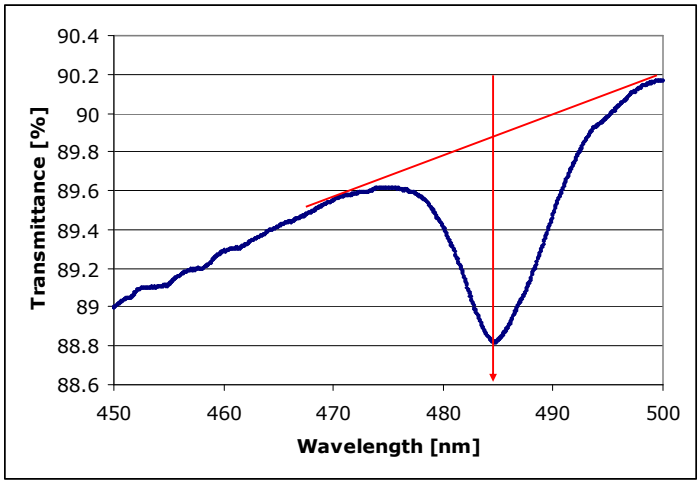

Fig. 2 : Transmittance of glass doped with a Tb^{3+} concentration of $1 \cdot 10^{20}$ cm^{-3} and a thickness of 1 cm in the range of 450 nm to 500 nm.

Evaluating the peak in Fig. 2 and setting the background to T=100 %, results in a transmittance in the peak of 98.78 %. With

$$T = e^{-N \cdot \sigma_{max} \cdot L} \tag{1}$$

N = 10^{20} cm^{-3} and L = 1 cm, a maximum cross-section of $\sigma_{max} = 1.23 \cdot 10^{-22}$ cm^2 results. This is about a factor of 4 to 10 times smaller than the cross-section of Nd^{3+} in glasses [9]. All absorption lines above 2000 nm are hidden by the OH absorption in the host.

In the UV spectrum we located lines between 378 nm and 317 nm. The absorption spectrum above 26 000 cm^{-1} is very complex [10] and therefore not all detected UV lines could properly be identified (Fig. 3).

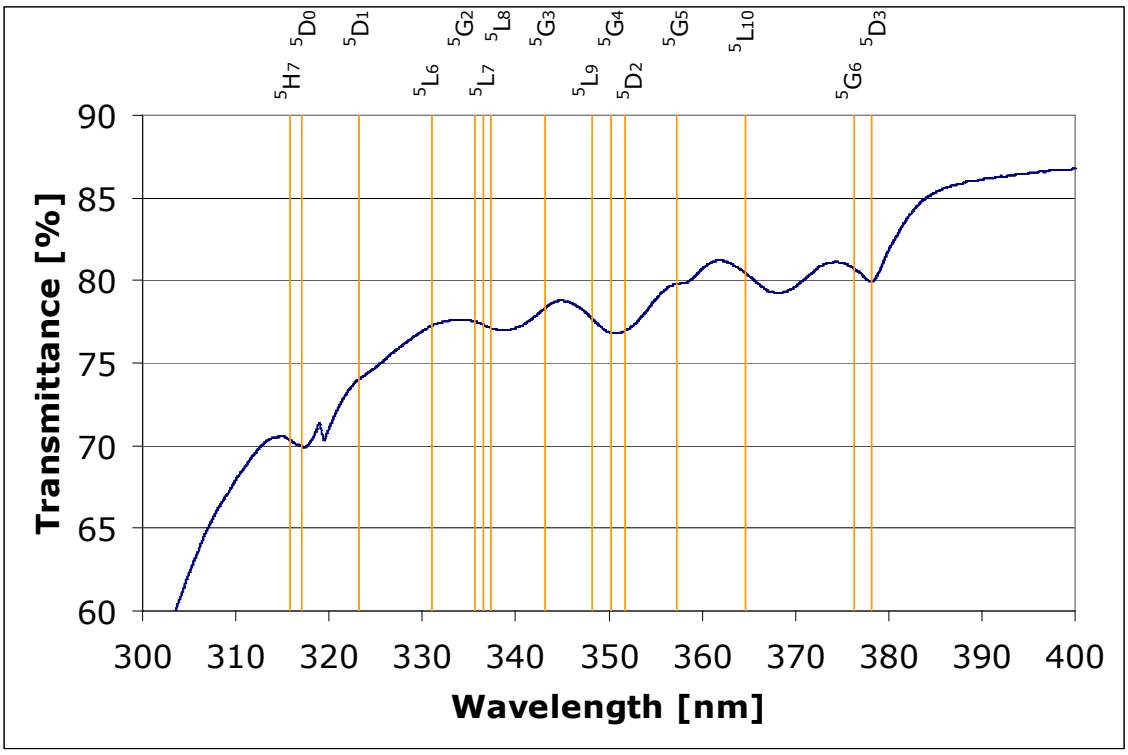

Fig. 3: Transmittance of glass doped with a Tb^{3+} concentration of $1\cdot 10^{20}$ cm^{-3} and a thickness of 1 cm in the range of 300 nm to 400 nm. The small peak at 320 nm is an artefact (change of detector) of the spectrometer.

The absorption at 317 nm is shown in more detail in Fig. 4:

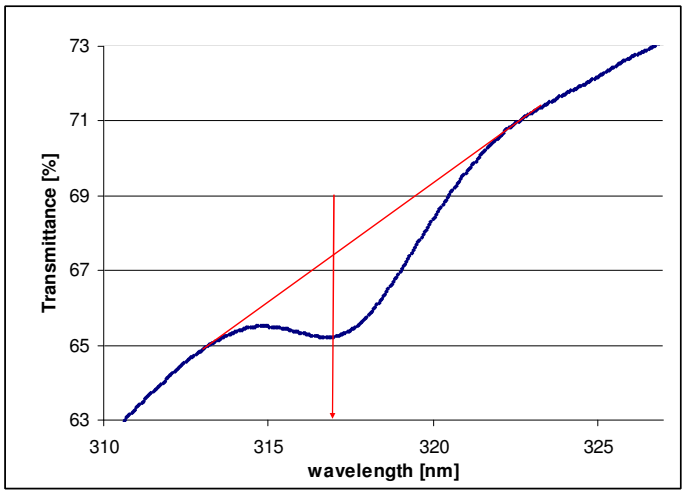

Fig. 4: Transmittance of glass doped with a Tb^{3+} concentration of $1\cdot 10^{20}$ cm^{-3} and a thickness of 1 cm in the range of 310 nm to 327 nm.

The measured absorption lines and their cross-sections are summarized in Tab. 2:

	λ measured [nm]	Transmittance [%]	$\Delta\lambda_{FWHM}$ [nm]	σ_{max} [cm^2]
$^7F_6 \rightarrow {}^7F_1$	1940	85.76	148	$1.54 \cdot 10^{-21}$
$^7F_6 \rightarrow {}^7F_0$	1885	84.77	230	$1.65 \cdot 10^{-21}$
$^7F_6 \rightarrow {}^5D_4$	485	98.91	8.7	$1.23 \cdot 10^{-22}$
$^7F_6 \rightarrow {}^5D_3$	378	93.41	8	$6.81 \cdot 10^{-22}$
?	368	92.67	10	$7.62 \cdot 10^{-22}$
$^7F_6 \rightarrow {}^5D_2$	358	95.16	5	$4.96 \cdot 10^{-22}$
?	350	93.61	10	$6.60 \cdot 10^{-22}$
?	338	96.54	8	$3.52 \cdot 10^{-22}$
?	317	97.33	4.2	$2.70 \cdot 10^{-22}$

Tab. 2: Measured absorption cross-sections in a glass doped with a Tb^{3+} concentration of $1 \cdot 10^{20}$ cm^{-3} and a thickness of 1 cm.

Based on the values of Tab. 2 the energy level diagram of Fig. 5 is found:

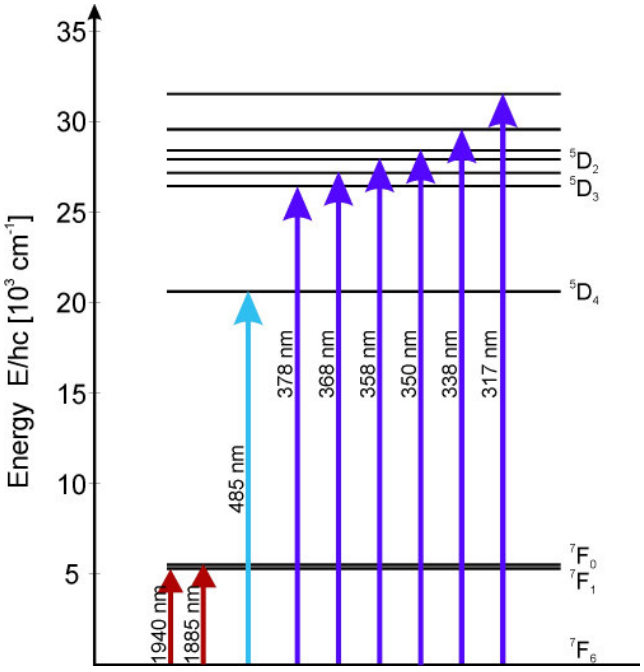

Fig. 5: Measured energy level diagram of Tb:glass

Emission spectroscopy with 488 nm excitation

Terbium has a strong fluorescence from the transition $^5D_4 \rightarrow ^7F_5$ in the green region around 542 nm. This fluorescence line is excited with 488 nm and measured as a function of pump intensity. If reabsorption of this line occurs, this would be visible as a saturation of the signal at high pump intensity. The chosen experimental arrangement is shown in Fig. 6:

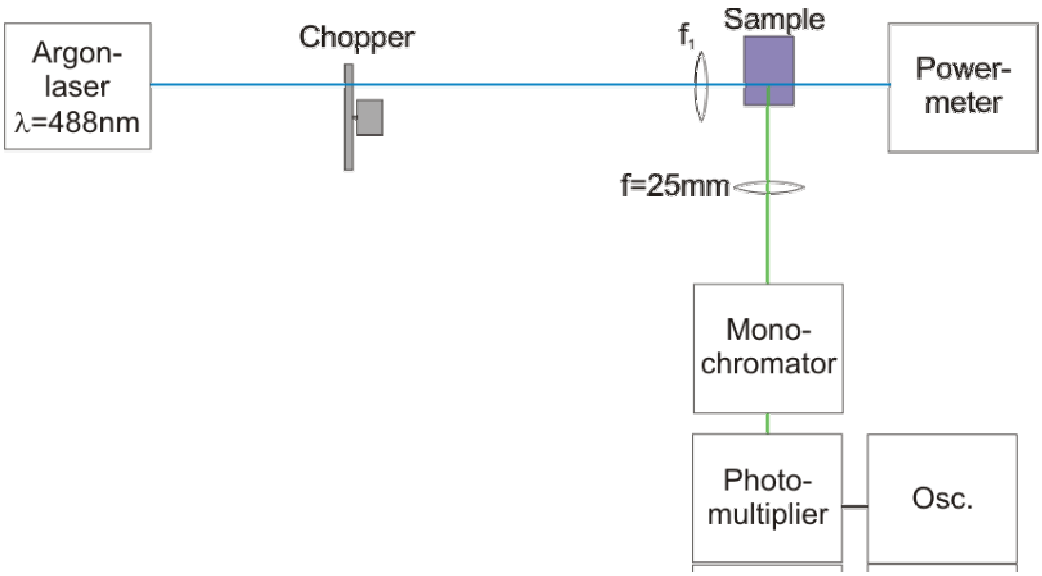

Fig. 6: Experimental arrangement for the measurement of the green fluorescence (λ = 542 nm) perpendicular to the laser beam.

The pump light is chopped with 37 Hz and a duty-cycle of 0.1. The pump power ranged up to about 350 mW leading to a peak intensity of 58 W/cm^2 without focussing, 50.2 kW/cm^2 when focussed with an f = 50 mm lens and 2.05 MW/cm^2 when focussed with a lens of f = 8 mm. In this range of intensity no deviation from a linear fluorescence output can be measured (Fig. 7).

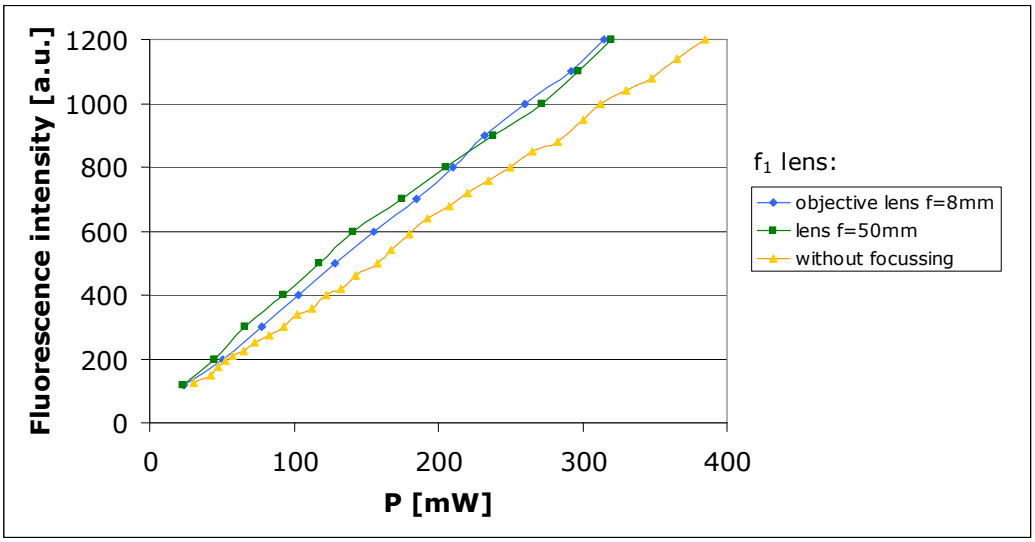

Fig. 7: Fluorescence intensity at λ = 542 nm. The higher fluorescence intensity with focussed pump beam is assigned to a better illumination of the monochromator slit.

To raise the sensitivity of the measurement the 542 nm fluorescence was also observed in the axis of the pump beam. Further, a 542 nm test light source, a bright luminescent diode (Distrelec L5-G81N-GT) in connection with a prism monochromator (Zeiss M4 QIII 40000), is used to detect possible amplification in the pumped glass sample. The green fluorescence is detected with a 20 cm grating monochromator, a photomultiplier (Hamamatsu R955) and a lock-in amplifier (Stanford Res. SR 530). Pump light @ 488 nm is shielded with a selective filter (Schott OG 515). The experimental arrangement is shown in Fig. 8.

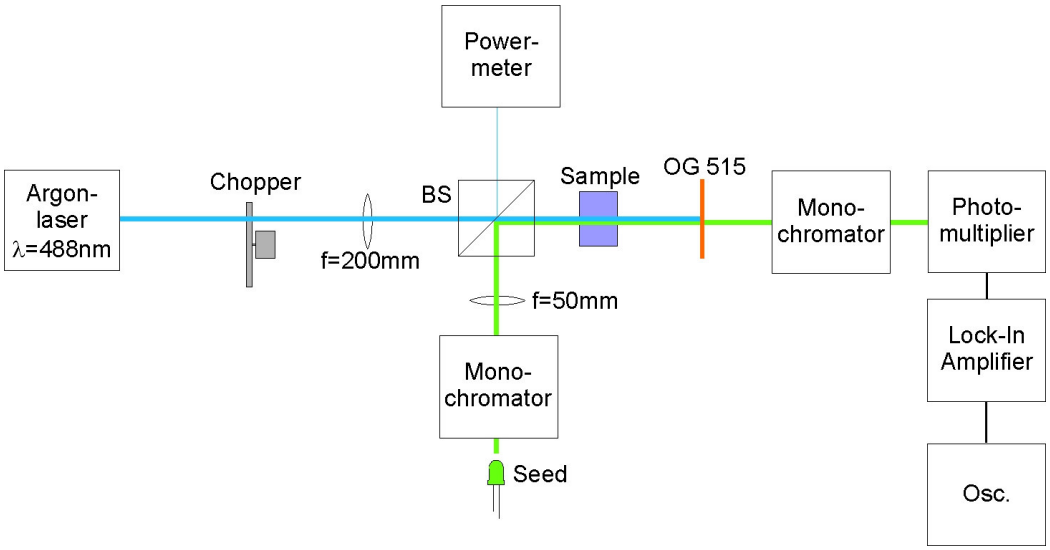

Fig. 8: Experimental arrangement. Measurement of the green fluorescence (λ = 542 nm) with a seed at λ = 542nm

The results are shown in Fig. 9:

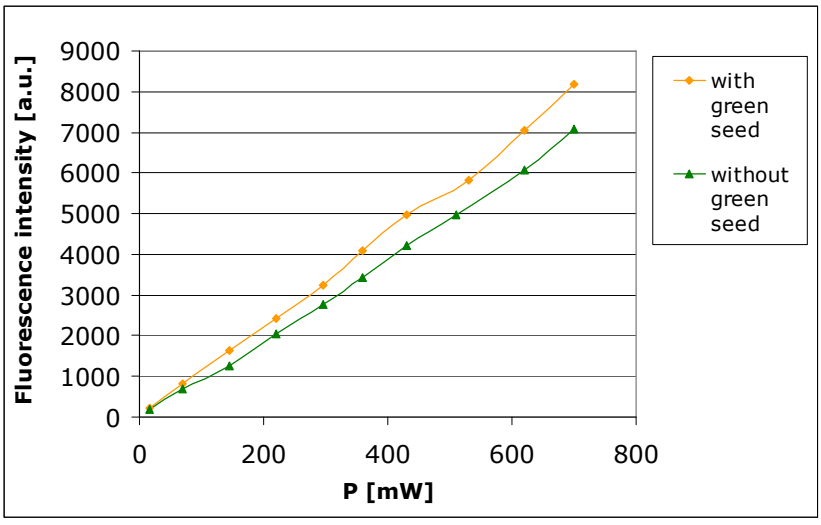

Fig. 9: Fluorescence intensity at λ = 542 nm. The laser beam is focussed with a f = 200 mm lens and the green seed with a f = 50 mm lens.

Also along the pump axis no deviation from linear, neither saturation nor amplification can be detected. Although the fluorescence in the sample is emitted in 4π and the detection system is placed 50 cm away from the sample, in all the measurements it was much stronger than the test light source.

UV fluorescence

Even if no deviation from linearity in the $^5D_4 \rightarrow ^7F_5$ transition is found, up-conversion due to excited-state-absorption or dipole-dipole interaction occurs. This is shown by the emission of UV-lines origination in the $^5D_3$ and higher levels. The UV- transitions measured in absorption have also been found in the fluorescence (Fig. 11). The experimental arrangement for the UV measurement is shown in Fig. 10:

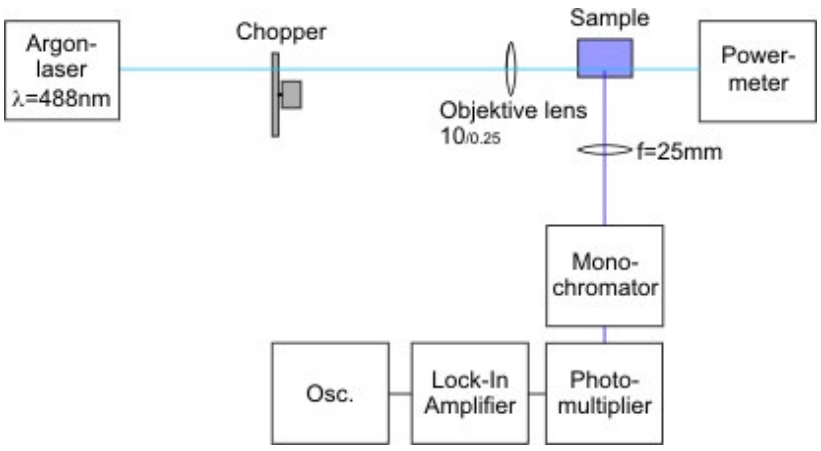

Fig. 10: Experimental arrangement for the measurement of the UV spectra.

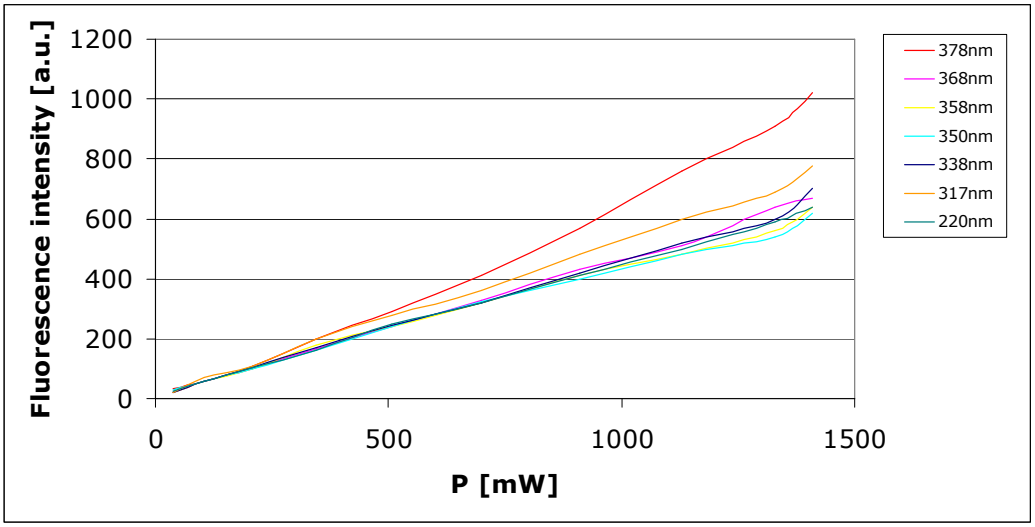

Fig. 11: Measurement of the UV fluorescence with the wavelength of λ = 378 nm, λ = 368 nm, λ = 358 nm, λ = 350 nm, λ = 338 nm, λ = 317 nm and λ = 220 nm

The emission of fluorescence with wavelengths below 488 nm originates from up-conversion-pumping. As a function of pump intensity, ESA as well as dipole-dipole interaction is a quadratic process and therefore a parabolic shape of the fluorescence intensity in Fig. 11 is expected. This is, however, not visible. We expect that the parabolic behaviour would be visible with much higher pump intensity and a longer light path in the material. Therefore it will be necessary to repeat the measurements in a Tb: silica fibre. These experiments are in progress.

Conclusion

We have measured absorption and emission spectra in Tb-doped Duran glass samples. The absorption cross-section of the $^7F_6 \rightarrow {}^5D_4$ pump transition has been determined to be $\sigma_{max} = 1.23 \cdot 10^{-22}$ cm^2. The intensity of the green fluorescence from $^5D_4 \rightarrow {}^7F_5$ has been measured as a function of 488 nm pump intensity. Within the range of intensity used in the experiment no deviation from linearity could be detected. Also in the intensity of UV-lines origination from up-conversion pumping measured as a function of pump intensity no quadratic characteristic could be detected.

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