ELSEVIER

Journal of Luminescence 60&61 (1994) 842–845

JOURNAL OF
LUMINESCENCE

Stimulated emission and excited-state absorption at room temperature on the 550 nm-laser transition in Er^{3+} doped YAlO_3

M. Pollnau^{*1}, E. Heumann, G. Huber*Institute of Laser-Physics, University of Hamburg, Jungiusstr. 11, D-20355 Hamburg, Germany*

Abstract

A pump- and probe-beam technique is used for measuring time-resolved and cw-pumped excited-state absorption (ESA) and stimulated-emission (SE) spectra of $\text{Er}^{3+}:\text{YAlO}_3$ with high resolution. In combination with absorption and fluorescence spectra, detailed information on the wavelengths and cross-sections of ESA and SE at the 550 nm laser transition is provided.

Laser action on the green Er^{3+} transition $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ at 550 nm has been achieved at low temperatures in several host materials, such as BaY_2F_8 [1], YAlO_3 [2], and LiYF_4 [3–5], and at room temperature in fluorozirconate fibres [6,7]. Recently room-temperature lasing in LiYF_4 has been demonstrated [8]. In several of these papers the possibility of parasitic ESA at 550 nm from the $^4\text{I}_{13/2}$ level has been discussed. CW-pumped ESA spectra of Er^{3+} in silica fibres [9] suggested that there may be an influence of this ESA on the laser performance at least in glasses and fibres. The exact wavelengths of lasing and ESA in crystals, however, depend on the spectral positions of single Stark transitions and on the corresponding cross-sections in the investigated host materials.

In this paper we present the measurements of cw-pumped and time-resolved ESA and SE in $\text{Er}:\text{YAlO}_3$ in the spectral region around 550 nm. Owing to the high resolution of 0.4 nm, single Stark transitions could be resolved. In combination with ground-state absorption (GSA) and fluorescence spectra, detailed information on the wavelengths and cross-sections of ESA and SE has been obtained.

A pump- and probe-beam technique is used for the measurement of ESA and SE spectra. The sample is excited at 488 nm by 100 μJ pulses of 10 ns duration from an excimer-laser-pumped dye laser (Coumarin 102). The Er ($2.9 \times 10^{20} \text{ cm}^{-3}$): YAlO_3 crystal (length $d = 9.6$ mm, crystallographic a -axis) is placed between two pinholes of 200 μm in diameter to define an excitation volume. The transmitted pump light is blocked by filters. Pulsed probe light from a broadband Xenon flash lamp is focused into the sample. The transmitted probe-light spectrum passes through a mono-

^{*} Corresponding author.

¹ Present address: Institute of Applied Physics, University of Bern, Sidlerstrasse 5, CH-3012 Bern, Switzerland.

chromator ($\frac{1}{4}$ m spectrometer with grating 1200 L/mm, resolution 0.4 nm) and is detected by an optical multichannel analyzer (OMA). A pulse and delay generator instantaneously triggers the excimer laser and, after an adjustable delay time, triggers the flash lamp and gates the OMA detector. The variation of the delay allows the measurement of time-resolved ESA and SE. The set-up for cw-pumped ESA and SE measurements is similar. An Ar⁺ laser excites the sample at 488 nm. A chopper of frequency 1 Hz and duty-cycle 1:1 provides the trigger signal and interrupts the Ar⁺ beam for the detection of the unpumped probe-beam spectra.

The probe beam has a low intensity compared to the pump beam, such that the laser-induced populations of the levels are unperturbed. The transmitted probe-beam intensity I_p after laser excitation and I_u without excitation as well as the cross-section σ_{GSA} are measured. The excited-state cross-section $\sigma_{\text{ESA}}(i)$ and $\sigma_{\text{SE}}(i)$ times the relative population factor n_i/n_e of the initial level i of the detected process at wavelength λ can then be calculated from the equation [10]

$$\frac{1}{n_e d} \ln(I_u/I_p) + \sigma_{\text{GSA}} = \sum_i \left\{ \frac{n_i}{n_e} [\sigma_{\text{ESA}}(i) - \sigma_{\text{SE}}(i)] \right\}. \quad (1)$$

The sum in Eq. (1) extends over all metastable and substantially populated levels. The time-resolved or cw excitation n_e is determined from the bleaching of the GSA in the $\ln(I_u/I_p)$ spectrum at wavelength regions where no ESA or SE occurs. The measurement of four spectra (application of no beam, pump beam, probe beam, pump and probe beam) results in a $\ln(I_u/I_p)$ spectrum, which is free of scattered pump light and spontaneous fluorescence from the sample [11]. Recently new possibilities of this measuring method have been demonstrated [12]. The temporal resolution of the ESA provides independent information on the absolute population densities n_i of the metastable levels as well as the effective excited-state cross-sections $\sigma_{\text{ESA}} - \sigma_{\text{SE}}$.

The GSA and fluorescence shown in Figs. 1(a) and 1(b) were monitored with a resolution of 0.10

Fig. 1. Comparison of (a) GSA $^4\text{I}_{15/2} \rightarrow ^4\text{S}_{3/2}$ (resolution 0.10 nm), (b) fluorescence $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ (resolution 0.08 nm) and (c) cw-pumped ESA $^4\text{I}_{13/2} \rightarrow ^2\text{H}_{9/2}$ (550–560 nm), $^4\text{I}_{11/2} \rightarrow ^2\text{G}_{7/2}$ and $^2\text{K}_{15/2}$ (560–570 nm) (resolution 0.4 nm) at 550 nm.

and 0.08 nm, respectively. The $^4\text{S}_{3/2}$ level consists of two Stark components. With the thermally coupled $^2\text{H}_{11/2}$ level taken into account, the Boltzmann populations of the $^4\text{S}_{3/2}$ Stark levels at 300 K are $b_{51} = 0.55$ and $b_{52} = 0.37$. The ground-state manifold consists of eight Stark components, which gives 16 transitions at 540–560 nm (Table 1). From the effective absorption cross-sections $\sigma_{\text{abs,eff}} = (\alpha/\text{doping concentration})$ the absolute atomic cross-sections σ are calculated under consideration of the Boltzmann populations. With the assumption of $\sigma_{\text{abs}}(\text{line } 9) = \sigma_{\text{em}}(\text{line } 9)$, the effective emission cross-sections $\sigma_{\text{em,eff}} = \sigma b_{5i}$ are calibrated. Line 9 is chosen because of a high cross-section with lower reabsorption losses than the lines 1 and 4.

Table 1

Absorption and emission cross-sections of the $^4I_{15/2} \leftrightarrow ^4S_{3/2}$ Stark transitions. ($\Delta\sigma_{\text{eff}} = \sigma_{\text{abs, eff}} - \sigma_{\text{em, eff}}$)

Transition $^4I_{15/2} \leftrightarrow ^4S_{3/2}$		λ [nm]	σ	Cross-section [10^{-20} cm^2]		
No.				$\sigma_{\text{abs, eff}}$	$\sigma_{\text{em, eff}}$	$\Delta\sigma_{\text{eff}}$
1	$1 \leftrightarrow 2$	540.8	5.1	1.6	1.9	-0.3
2	$2 \leftrightarrow 2$	542.3	3.1	0.7	1.3	-0.6
3	$1 \leftrightarrow 1$	543.2	1.5	0.4	1.0	-0.6
4	$2 \leftrightarrow 1$	544.7	5.5	1.4	2.9	-1.5
5	$3 \leftrightarrow 2$	545.8	1.7	0.2	0.6	-0.4
6	$4 \leftrightarrow 2$	547.3	3.2	0.3	1.3	-1.0
7	$3 \leftrightarrow 1$	548.3	0.9	0.1	0.5	-0.4
8	$5 \leftrightarrow 2$	548.8	1.7	0.2	0.6	-0.4
9	$4 \leftrightarrow 1$	549.7	3.7	0.4	2.0	-1.6
10	$5 \leftrightarrow 1$	551.2	1.6	0.1	0.9	-0.8
11	$6 \leftrightarrow 2$	552.5	2.6	0.1	0.9	-0.8
12	$7 \leftrightarrow 2$	554.2	1.1	0.1	0.3	-0.2
13	$6 \leftrightarrow 1$	554.9	0.4	0.0	0.2	-0.2
14	$8 \leftrightarrow 2$	556.7	?	0.0	0.4	-0.4
15	$7 \leftrightarrow 1$	556.7				
16	$8 \leftrightarrow 1$	559.2	0.4	0.0	0.2	-0.2

The absolute cross-sections are then taken as an average of the values determined from absorption and emission spectra (Table 1). All values of the difference $\Delta\sigma_{\text{eff}}$ of the effective cross-sections are negative, which means population inversion on all Stark transitions, if an equal population of the $^4S_{3/2}$ and $^4I_{15/2}$ levels is achieved. The highest value of $\Delta\sigma_{\text{eff}}$ is calculated for the laser line at 549.7 nm [2], which will be the possible transition for pulsed laser action at room temperature, too. Its terminating Stark level has a Boltzmann population $h_{04} = 0.11$.

Stimulated emission at room temperature is detected up to 80 μs after pulsed excitation, see Fig. 2(a). A calculation of Eq. (1) considering the Boltzmann population ($n_i = n_5 h_{51}$) leads to the following result for the 549.7 nm transition: with the measured quantity (left-hand side of Eq. (1)) at 549.7 nm being $1.2 \times 10^{-20} \text{ cm}^2$ (Fig. 2(a)), $n_5/n_e = N_5 = 0.68$ at 8 μs delay [10], and $h_{51} = 0.55$, a cross-section $\sigma = 3.2 \times 10^{-20} \text{ cm}^2$ is determined from SE, in excellent agreement with the value $\sigma = 3.7 \times 10^{-20} \text{ cm}^2$ derived from GSA (Table 1). Therefore, this measuring method will be suitable for the precise determination of cross-sections between two excited states, e.g. the laser transition

Fig. 2. Time-resolved ESA and SE at 550 nm, delay (a) 8 μs , (b) 200 μs , and (c) 2 ms (resolution 0.4 nm).

of a four-level laser or ESA. Population inversion at 549.7 nm

$$h_{51}N_5n_e > h_{04}(1 - n_e) \Rightarrow n_e > 20\% \quad (2)$$

requires short-pulse excitation above 20% into the $^4F_{7/2}$ or $^2H_{11/2}$ level, which is a high request to available pump lasers.

At 200 μs after pulsed excitation the relative population n_i/n_e of the $^4I_{11/2}$ level reaches its maximum and strong ESA at 560–570 nm on the transition $^4I_{11/2} \rightarrow ^2G_{7/2}$ and $^2K_{15/2}$ is detected, see Fig. 2(b). It does not affect the fluorescence, because there is no spectral overlap. At long delay times the remaining excitation concentrates in the $^4I_{13/2}$ level and leads to the ESA transition

$^4I_{13/2} \rightarrow ^2H_{9/2}$ (Fig. 2(c)). This ESA influences the long wavelength side of the green fluorescence and counteracts SE even at short delay times (Fig. 2(a)). The short wavelength end of this ESA lies only a few angström away from the lasing line at 549.7 nm.

There is no SE detected when the crystal is cw pumped. Transitions terminating at high Stark levels of the ground state, which have low Boltzmann populations, are heavily influenced by the cw ESA $^4I_{13/2} \rightarrow ^2H_{9/2}$ (see Fig. 1(c)) and, therefore, will not exhibit cw-pumped SE in any case. We achieved quenching of this ESA by co-doping of an $\text{Er}(2.0 \times 10^{20} \text{ cm}^{-3}) : \text{YAlO}_3$ with $\text{Tb}(0.2 \times 10^{20} \text{ cm}^{-3})$. The energy transfer $\text{Er } ^4I_{13/2} \rightarrow \text{Tb } ^7F_0$ reduces the $^4I_{13/2}$ lifetime and, hence, the ESA to 56% (4.3 ms instead of 7.7 ms in $\text{Er}(2.9 \times 10^{20} \text{ cm}^{-3}) : \text{YAlO}_3$), whereas the $^4S_{3/2}$ lifetime remains nearly unchanged (93 μs versus 102 μs).

In conclusion, we measured time-resolved and cw-pumped excited-state absorption and stimulated emission in $\text{Er}^{3+} : \text{YAlO}_3$ at 550 nm with a spectral resolution of 0.4 nm. The stimulated emission cross-sections derived from these measurements are in good agreement with those obtained from absorption and fluorescence measurements, which introduces a new measuring method for the precise determination of absolute cross-sections between two excited states. Stimulated emission at room temperature on the $^4S_{3/2} \rightarrow ^4I_{15/2}$ fluorescence at 550 nm exists up to 80 μs after pulsed excitation.

Acknowledgements

We wish to thank H. Stange and T. Danger of the Institute of Laser-Physics, University of Hamburg, Germany, for the development of most parts of the experimental arrangement. This work was supported by the Deutsche Forschungsgemeinschaft.

References

- [1] L.F. Johnson and H.J. Guggenheim, *Appl. Phys. Lett.* 20 (1972) 474.
- [2] A.J. Silversmith, W. Lenth and R.M. Macfarlane, *J. Opt. Soc. Am. A* 3 (1986) P128; *Appl. Phys. Lett.* 51 (1987) 1977.
- [3] W. Lenth, R.M. Macfarlane and A.J. Silversmith, *Opt. News* 13 (1987) 26.
- [4] R.A. McFarlane, *Appl. Phys. Lett.* 54 (1989) 2301.
- [5] P. Xie and S.C. Rand, *Opt. Lett.* 17 (1992) 1116; *Opt. Lett.* 17 (1992) 1198.
- [6] T.J. Whitley, C.A. Millar, R. Wyatt, M.C. Brierley and D. Szebesta, *Electron. Lett.* 27 (1991) 1785.
- [7] J.Y. Allen, M. Monerie and H. Poignant, *Electron. Lett.* 28 (1992) 111.
- [8] R. Brede, T. Danger, E. Heumann, G. Huber and B. Chai, *Appl. Phys. Lett.*, 63 (1993) 729.
- [9] R.I. Laming, S.B. Poole and E.J. Tarbox, *Opt. Lett.* 13 (1988) 1084.
- [10] M. Pollnau, E. Heumann and G. Huber, *Appl. Phys. A* 54 (1992) 404.
- [11] S.A. Payne, L.L. Chase and G.D. Wilke, *Phys. Rev. B* 37 (1988) 998.
- [12] M. Pollnau, E. Heumann and G. Huber, *Phys. Stat. Sol. A* 130 (1992) K121.