Characterising Energy Transfer Upconversion in Nd:YVO_4 at Elevated Temperatures

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Abstract: Energy Transfer Upconversion and $^4F_{3/2}$ energy level absorption cross section are measured in Nd:YVO_4 at temperatures ranging from 300 K to 450 K. The ETU coefficient decreases from $(34.5 \pm 6.5) \cdot 10^{-17}\ cm^3/s$ to $(3.0 \pm 2.0) \cdot 10^{-17}\ cm^3/s$.

OCIS codes: (190.7220) Upconversion, (140.3530) Lasers, neodymium, (300.1030) Absorption.

1. Introduction

Nd-doped vanadate crystals are among those commonly used in diode-pumped solid state lasers. Compared to Nd:YAG, both Nd:YVO_4 and Nd:$GdVO_4$ have higher absorption cross sections around 808 nm (π-pol) which, combined with a large emission cross section, provide high gain with short crystal lengths. Both crystals are typically employed in solid-state lasers operating on the 1.06 μm emission line, however, the lower gain 0.9 μm transition could potentially be more efficient due to a lower quantum defect between pump and output wavelengths. As the latter transition suffers reabsorption losses and it has lower emission cross section, it is susceptible to the detrimental thermal effects associated with the waste heat deposited during the excitation processes. As such its laser performance could be improved if key contributors to these effects are well characterised.

Parasitic effects like Energy Transfer Upconversion (ETU) and Excited State Absorption (ESA) are deleterious for transitions that require high-irradiance pumping, such as the quasi-four-level Nd^{3+} system, as they reduce the relatively low available gain. ETU de-excites the upper laser level through ion-ion interaction, while ESA anihilates pump (or signal) photons, and both excite ions from the $^4F_{3/2}$ level to higher energy states. Waste heat is produced via the non-radiative decay channels of these excited ions.

In this work, we present a comprehensive study of the $^4F_{3/2}$ energy level absorption cross section over the range from 300 K to 450 K. These results, which show a decrease in cross section with increasing temperature, were key to the characterisation of the macroscopic ETU parameter over the same range of temperatures. By means of a z-scan technique, we measured the ETU coefficient for Nd:YVO_4 with the pump laser tuned to the strongest 808 nm σ-polarisation peak: it was found to decrease from $(125 \pm 15) \cdot 10^{-17}\ cm^3/s$, compatible with our previous result at room temperature [2], to $(9.75 \pm 2.75) \cdot 10^{-17}\ cm^3/s$. However, with the pump tuned to 806 nm, the second strongest σ-polarisation absorption peak, the maximum ETU parameter obtained was $(34.5 \pm 6.5) \cdot 10^{-17}\ cm^3/s$. It appears that an additional ESA loss process occurs for the 808 nm σ-polarised pump, in the vicinity of room temperature, which corresponds to a phonon ($\hbar\omega = 155\ cm^{-1}$) assisted transition that coincides with a vibrational mode of the crystal [4].

This information, coupled with elevated temperature emission cross section data [3], provide a new level of detail for the design parameters needed for these materials, which may have direct impact on further optimisation of vanadate lasers operating at elevated temperatures or under intense pumping conditions.

2. Methodology

2.1. Absorption cross section measurements

Small signal absorption measurements, well described by the Beer-Lambert law, can provide an accurate measure of the absorption cross section, once the total number of doping ions N_{tot} and the length of the crystal L are known:

$$\sigma_{abs}(\lambda) = \frac{\ln(\frac{I_{in}(\lambda)}{I_{out}(\lambda)})}{N_{tot} L} \tag{1}$$

We recorded the input and the transmission spectra of the broadband amplified spontaneous emission (ASE) of a sub-threshold fibre-coupled diode-laser. Accounting for Fresnel reflections at the uncoated facets of the 1 mm long, 1$at.\%$-doped Nd:YVO_4 crystal, whilst taking into account the probe polarisation, eqn. (1) was calculated.

We characterised the absorption cross section from 770 *nm* to 840 *nm* at several different temperatures in the range 300 *K* to 450 *K*. The details of the experimental setup can be found in our previous paper [1].

2.2. ETU coefficient measurements

Employing our z-scan experimental setup [1], we measured the transmission of a focussed Ti:sapphire laser beam as a function of z-position in the same temperature range explored for the absorption cross section measurements, and compared it to the 2-level spatially dependent rate equation model described in [1]. Furthermore, to account for the discrepancy observed between the two pump wavelengths, the equation for the longitudinal evolution of the pump irradiance $I_p(r,z)$ was modified as in eqn. (2) in order to include a pump ESA process.

$$\frac{dI_p(r,z)}{dz} = I_p(r,z)(-\sigma_{abs}N_1(r,z) - \sigma_{ESA}N_2(r,z)) \tag{2}$$

N_1 and N_2 are the population densities of the ground and excited states respectively, σ_{abs} is the measured ground state absorption cross section, σ_{ESA} is the absorption cross section from ${}^4F_{3/2}$ to ${}^2D_{5/2}$ level , $h\nu_p$ is the pump photon energy. In addition the macroscopic ETU coefficient , W_{ETU}, is needed in the rate equations.

At the point of highest pump irradiance the pump transmission is significantly lower than expected from the saturation intensity alone: this is due to ETU (and/or ESA) countering ground state bleaching.

Once all the other modelling parameters were fixed through measurement, i.e. the absorption cross section, or taken from existing literature, a tolerance band around the simulated curve was defined and our experimental data was fitted within this band by varying the ETU coefficient, the only remaining fitting parameter in the case of negligible ESA.

3. Experimental results and discussion

3.1. Absorption cross section measurements

As data shows in fig. 1 and 2, the highest peak for the π-polarisation absorption cross section decreased from 58.6 ± 0.2 pm^2 at RT to 30.9 ± 0.6 pm^2 at 450 *K* and red-shifted 0.2 *nm* at the highest temperature. The maximum value for the σ-polarised spectrum reduced from 12.4 ± 0.2 pm^2 to 6.57 ± 0.15 pm^2, also red-shifting by 0.2 *nm* over the whole temperature range.

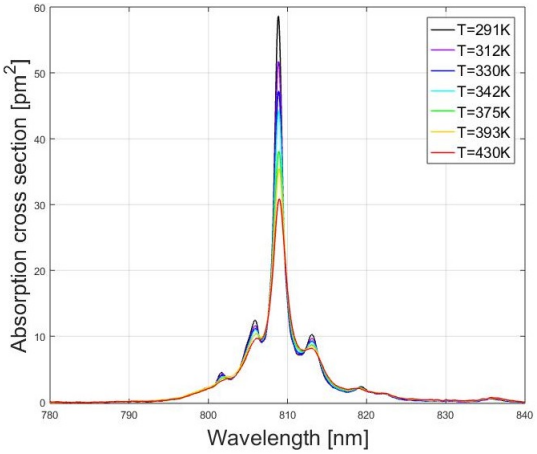

Fig. 1. Nd:YVO_4: π-pol absorption cross section.

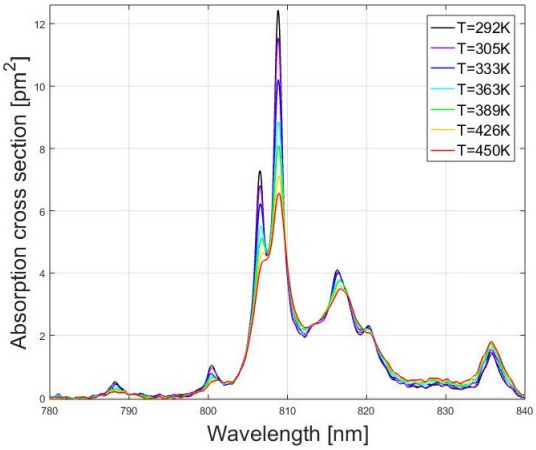

Fig. 2. Nd:YVO_4: σ-pol absorption cross section.

3.2. ETU coefficient measurements

We tuned our Ti:sapphire laser to both stronger σ-polarisations absorption peaks in order to probe the irradiance dependent transmission. Its beam quality was $M^2 = 1.05$ in both axes with second moment beam waist radii $w_x = 19.7 \pm 0.2$ μm and $w_y = 19.9 \pm 0.2$ μm after a moveable 200 *mm* focussing lens, providing an average confocal parameter of 3.05 ± 0.15 *mm*, thus satisfying our assumption that the beam doesn't change significantly through the sample. Given a 220 mW input power, the maximum available irradiance was 36 kW/cm^2, almost twice the saturation

irradiance for the 808 *nm* pump, and comparable to that for 806 *nm*. The fluorescence lifetime of the ${}^4F_{3/2}$ level in the small-signal regime was constantly monitored throughout the whole temperature range and showed no change, providing an average value of $93.1 \pm 0.2\ \mu s$.

The distinctly different trend in the ETU dependence on temperature for the two different pump wavelengths raised the question of whether there was another process involved, which was not catered for in model [1]. A possible mechanism for reducing the transmission of the pump power with increasing irradiance is, as illustrated in eqn. (2), ESA. Possible values for the ETU coefficient and ESA cross section for σ-polarisation of the 808 *nm* pump are listed in table 1. Fig. 3 and 4 show the comparison of experimental z-scan data with our theoretical model and the ETU coefficient variation with temperature, respectively.

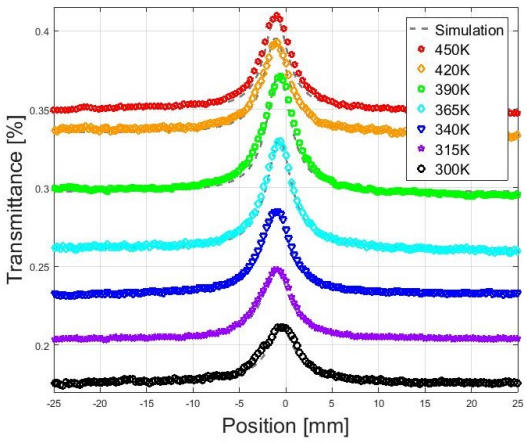

Fig. 3. 1*at*.% Nd:YVO_4: z-scan data vs theoretical model.

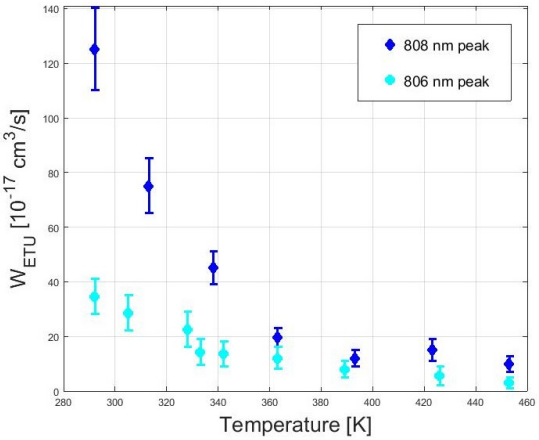

Fig. 4. 1*at*.% Nd:YVO_4: W_{ETU} vs Temperature.

T [K]	$W_{ETU}^{(808nm)}$ $[10^{-17}\ cm^3/s]$	$W_{ETU}^{(806nm)}$ $[10^{-17}\ cm^3/s]$	$W_{ETU}^{(808nm_{ESA})}$ $[10^{-17}\ cm^3/s]$	σ_{ESA} $[pm^2]$
300	125.0 ± 15.0	34.5 ± 6.5	33.0 ± 5.0	4.2 ± 0.3
325	75.0 ± 10.0	28.5 ± 6.5	27.5 ± 4.5	2.8 ± 0.3
350	45.0 ± 6.0	14.2 ± 4.7	23.0 ± 4.0	1.5 ± 0.3

Table 1.

4. Conclusions

In conclusion, we have extensively characterised the ${}^4F_{3/2}$ Nd:YVO_4 absorption cross section for different elevated temperatures through simple absorption measurements and determined the variation in ETU coefficient in the same range employing the z-scan technique. Experimental data shows possible ESA for the σ-polarised 808 *nm* pump: further investigation on the subject is in progress. We will report on similar measurements with Nd:$GdVO_4$.

References

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