# International Conference on Space Optics—ICSO 2004

Toulouse, France 30 March–2 April 2004

Edited by Josiane Costeraste and Errico Armandillo



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International Conference on Space Optics — ICSO 2004, edited by Errico Armandillo, Josiane Costeraste, Proc. of SPIE Vol. 10568, 105681B · © 2004 ESA and CNES CCC code: 0277-786X/17/\$18 · doi: 10.1117/12.2307963

## Mixed Garnet Laser Crystals for Water Vapour DIAL Transmitter

Rainer Treichel<sup>(1)</sup>, Christoph Czeranowsky<sup>(2)</sup>, Bilge Ileri<sup>(2)</sup>, Klaus Petermann<sup>(2)</sup>, Günter Huber<sup>(2)</sup>

<sup>(1)</sup> EADS ASTRIUM GmbH, 88039 Friedrichshafen, German, E-mail: rainer.treichel@astrium.eads.net
<sup>(2)</sup> University of Hamburg Luruper Chaussee 149, 22761 Hamburg, Germany, E-mail: czeranowsky@physnet.uni-hamburg.de

### ABSTRACT

There are more or less well established technologies such as the optical-parametric-oscillator (OPO), the Raman-laser, and the Ti-Sapphire laser, which are able to emit laser light in the region of the water vapour absorption lines. For WALES the regions of about 935 nm, 942 nm, and 944 nm have been identified as the most suitable wavelength ranges. However, each of these laser designs is highly sophisticated. Current baseline for WALES is the Ti-Sapphire laser.

A fourth possibility to achieve these wavelength ranges is to shift the groundstate laser lines (938 nm and 946 nm) of the Nd:YAG laser by replacing Aluminium and Yttrium by other rare earth elements. Changes of the host lattice characteristics lead to a shift of the upper and lower laser levels. These modified crystals are summarized under the name of "Mixed Garnet" crystals. Only the Mixed Garnet lasers can be pumped directly with diode laser and use a direct approach to generate the required laser pulses without frequency conversion. Therefore no additional non-linear crystals are needed and a higher electric to optical efficiency is expected as well as single frequency operation using spectral tuning elements like etalons. Such lasers have the great potential to fulfil the requirements and to become the preferred transmitter concept for WALES as well as for follow up missions. Within a ESA study several crystal compositions have been grown, spectrally characterised and analysed.

Absorbed space radiation energy in the crystal lattice causes colour centres, which can reabsorb the pump and laser wavelength and consequently reduce the laser gain considerably. Co-dopants such as Chromium and Cerium are able to suppress the colour centres and are candidates for effective radiation hardening.

The results of the crystal tuning, the co-doping with different radiation hardeners and the radiation tests will be presented. There applicability for a space based water vapour DIAL transmitter will be discussed.

## 1. INTRODUCTION

The Nd<sup>3+</sup>-ion offers various groups of laser lines in the near IR spectral region. There are transitions from the

 ${}^{4}F_{3/2}$  upper laser level into the  ${}^{4}I_{13/2}$ ,  ${}^{4}I_{11/2}$ , and  ${}^{4}I_{9/2}$ manifold. In Nd:YAG the strong laser transition into the  ${}^{4}I_{9/2}$  is located at 946 nm. The wavelengths are shifted to shorter values by enlarging the lattice constant. This is caused by the following three effects: 1. as the main effect, the crystal field and therefore the Stark splitting and the emission wavelength is decreased,

2. due to the decreasing LS-coupling, the splitting between the manifolds of the  ${}^{4}I$  (and  ${}^{4}F$ ) and therefore the wavelength increases,

3. further more the smaller Coulomb interaction causes a reduction of the splitting between the <sup>4</sup>F and the <sup>4</sup>I level, leading to a longer wavelength.

Enlarging the lattice constant and thus decreasing the crystal field strength can easily be realized in the garnet system  $RE_3M_5O_{12}$ , where RE = Y, Lu, Gd, and mixtures thereof, M = Al, Ga, Sc, and mixtures thereof, and  $Nd^{3+}$  is entering the RE-site.

# WATER VAPOUR DIAL WAVELENGTHS AND DIAL PRINCIPLE

The main objective of the WALES mission is to provide essential information on the distribution of atmospheric water vapour and aerosols in the troposphere and lower stratosphere (from 0 to 16 km altitude) for numerical weather prediction, climatology and atmospheric modelling. Absolute quantitative measurements of water vapour will be obtained over a large dynamic range (from 0.01 to 15 g/kg), with a high level of vertical resolution and accuracy not achievable by other systems. The spatial and temporal variation of water vapour will be directly sampled with global coverage, not only above and below optically thin clouds but also above cloud tops and in cloud gaps between dense clouds. The independent set of global water vapour profiles can be used to validate other measuring techniques such as operational passive infrared and microwave. Secondary informations can also be derived such as cloud tops and base heights, optical thicknesses, planetary boundary layer height, and surface reflectance/albedo.

In the WALES Phase A study three wavelength regions each with a suitable subset of four wavelengths have each been identified. The preselection of these 3 times 4 wavelengths (see Table 1) is a result of a trade-off, which presents the best compromise between scientific requirements and technical realizability.

*Table 1: Absorbtion wavelength sets for Water Vapour Dial* 

Set		Line	$\lambda_{o}$
		Strong	935.684 nm
935	nm	Medium	935.561 nm
group		Low	935.906 nm
		Offline	935.4 nm
		Strong	943.0822 nm
942 group	nm	Medium	942.441 nm
		Low	943.247 nm
		Offline	940.0 nm
			(942.7 nm TBC)
		Strong	944.367 nm
944	nm	Medium	944.858 nm
group		Low	945.235 nm
		Offline	945.0 nm

The nadir-pointing DIAL instrument performs differential measurements of absorption at four closely separated wavelengths, three of which are 'online' measurements at molecular absorption lines of water, and one 'off-line' measurement, where absorption is significantly reduced. In this way, the different subintervals of the dynamic absorbtion range are addressed by dedicated on-line wavelengths. The different on-line wavelengths possess different penetration depths, thus allowing measurements over different altitude intervals. The strongly absorbing water vapour lines are used for higher altitudes (low water vapour concentration) and the weakly absorbing lines are used for lower altitudes (high water vapour concentration). The on-line measurements and the separate off-line measurements are used in a cascaded way, i.e. for each on-line measurement, the measurement on the next weaker line serves as its offline measurement.

#### 2. TUNING STRATEGY

The wavelengths required for the water vapour dial can not be generated by standard Neodymium lasers. Although the 946 nm emission of Nd:YAG enables a first approach, mixed garnets have to be used to substitute the laser gain material. Mixed garnets generate the required wavelengths by direct transitions between the  ${}^{4}F_{3/2}$  and  ${}^{4}I_{9/2}$  manifold of the Neodymium ion (see Figure 1).

The spectra shown in Figure 2 give –as an example- a plot of the Neodymium emission in the region of 940 nm in various mixed garnets compared to

Nd:YAG. They make clear that the admixture of YSGG does slightly shift the spectral emission. Assuming that the emission cross section is affordable high, laser operation at the needed water vapour lines seems possible. A composition of e.g. 59% YAG and 41% YSGG is near optimum to generate the required water vapour absorption lines around  $\lambda = 943$  nm. Although the presented spectra show a double peak, it should be noticed that due to the smaller cross section and strong competition to the major peak efficient operation on the minor emission peak can not be expected. For the shorter water vapour emission region around 935 nm it is therefore necessary to focus on the mainly peak.



*Fig 1: : Level Scheme and transition of*  $Nd^{3+}$ : *YAG* 

The strategy was to tune the garnet system  $RE_3M_5O_{12}$ , where RE = Y, Lu, Gd, and mixtures thereof, M = Al, Ga, Sc, and mixtures thereof.  $Nd^{3+}$  is, entering the REsite:

	LuAG	$Lu_3Al_5O_{12}$
	YAG :	$Y_3Al_5O_{12}$
	YSAG :	Y <sub>3</sub> (Sc,Al) <sub>5</sub> O <sub>12</sub>
	LuGG:	Lu <sub>3</sub> Ga <sub>5</sub> O <sub>12</sub>
YGG :	Y <sub>3</sub> Ga <sub>5</sub> O	12
	YSGG :	$Y_3(Sc,Ga)_5O_{12}$
	GSAG :	$Gd_3(Sc,Al)_5O_{12}$
	GGG:	Gd <sub>3</sub> Ga <sub>5</sub> O <sub>12</sub>
	GSGG:	$Gd_3(Sc,Ga)_5O_{12}$
	LaLuGG:	$La_3(Lu,Ga)_5O_{12}$ .

From the top (LuAG, YAG) to the bottom (GSGG) bigger ions are introduced into the crystal lattice increasing the lattice constant and decreasing the strength of the crystal field compared to YAG. As a main consequence the ground state Stark splitting of

the  ${}^{4}I_{9/2}$ -level decreases and tunes therefore the 946 nm laser transition of Nd:YAG to shorter wavelengths.



*Fig.2:* Nd<sup>3+</sup> ground state transitions of some selected crystals in the spectral region of interest. [F.S. Ermeneux et al., OSA TOPS Vol. 26, 242 (1999)]

#### 3. GROWN COMPOSITIONS

To find suitable garnets for the required wavelengths around 935 nm, 942 nm, and 944 nm the following compositions have been grown and spectroscopically investigated:

YSAG LaLuGG GSGG GSAG YGG LuGG

In contrast to Barnes et all the aim of this work is to find crystals that have been made up of only two or three different ions.

#### 3.1. YSAG

In the YSAG composition the Aluminium ions are replaced partially by Scandium ions. Depending on the mixing ratio of the compounds the emission wavelength is more or less shifted. Figure 3 shows the emission spectrum of three different Scandium concentrations, i. e. about 10%, 20%, and 28%. With the highest concentration of 28% Scandium the major peak has been shifted from 946 to near 944 nm.



Fig. 3: Emission Spectra of YSAG with tree different Scandium concentrations

Although the wavelength group around 944 nm has been achieved with the 28% Scandium concentration, the crystal quality and stability are very pure. Due to the larger ion size of Scandium a considerable deformation of the lattice leads to extreme stress in the crystal and cracks cannot be avoided anymore. Because of the non-congruent melting behaviour this garnet was found to be less suitable for growing large crystals and therefore was not further investigated.

#### 3.2. LALUGG

The composition of Lanthanum, Lutetium, and Gallium has already very large ions, which form a lattice with an extreme large constant. Both emission peaks are therefore shifted below 935 nm and 924 nm. Here, the replacement of Lanthanum by Lutetium decreases the lattice constant and induces a shift to longer wavelengths. However in Figure 4 it can be seen, that



*Fig. 4: Emission spectra of several Nd-doped LaLuGG compositions* 

the shift of the main fluorescence peak is relatively weak. The wavelength group around 935 nm was not achieved even with the highest Lu concentration.

#### **3.3. GSGG**

In comparison to YSAG the Yttrium is replaced in this garnet by the bigger Gadolinium. Therefore the crystal lattice is increased and the emission is shifted to shorter wavelength. Additionally the Scandium fits better into the larger crystal and causes less stress than in the YSAG. The aluminium is replaced by the bigger Gallium, which decreases further the emission wavelength. However it can be seen in Figure 5 that GSGG has the shortest emission wavelength at 936.6 nm, which is not sufficient for the 935 nm-group.

### 3.4. GSAG

The GSAG emission is shorten than the one of YSAG due to the bigger Gadolinium instead of Yttrium, but longer than in GSGG, because the aluminium is smaller than Gallium. The stronger emission peak hits the 942 nm group nearly perfect and due to the Gadolinium the Scandium fits better into the crystal and causes less stress. So, crystals of good quality should be achievable. Interesting is the large separation of the two emission peaks in GSAG.

#### 3.5. YGG, LUGG

YGG and LuGG are shifted to shorter emission wavelengths by replacing the Aluminium by Gallium. Surprising is the switching of the height of the two emission peaks: in these two crystals the shorter emission peak is the stronger one. This strong emission peak in YGG hits very good the water vapour absorption lines around 935 nm.



Fig. 5: Emission spectra of GSAG, LuGG, YGG, and GSGG.

#### 4. RADIATION TESTS

Nd:YAG is known to get high radiation hardness when it is co-doped with  $Cr^{3+}$ . Also Ce co-doping increases the radiation hardness.

Therefore in a first step YSAG and GSGG have been co-doped with Cr and Ce and have been irradiated with Gamma radiation at expected doses of a typical WALES mission (sun-synchronous orbit, 450 km altitude, 3 years mission duration).

The calculated dose amounts to 1.4 krad. By comparison of the absorption spectra of the crystals before and after irradiation the possible appearance of colour centres should be detected.

As expected the relative low dose did not change any of the absorption spectra. In a planned second step a factor five or ten higher dose will be put on a second set of samples. The third step is to irradiate the samples with protons. This work is still going on.

#### 5. CONCLUSION

The most promising compositions for reaching the water vapour absorption lines are YGG and GSAG. YGG emits with its shorter emission peak at 935 nm, which exhibits a higher cross section than the longer emission peak.

GSAG is useful for the 942 nm wavelength group. So, further work is now concentrated on these two systems. Further more, both garnets are known to posses a congruent melting behaviour, which makes the growth of large crystal boles less complicated.

#### 6. ACKNOWLEDGEMENT

This work is being done under the ESA contract no. 17160/03/NL/JA.