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Ce^{3+} Sensitized Emission of Nd^{3+} in Garnet Structures

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Introduction

This work is part of the research project CoMaMed – Converter Materials for Laser Diodes in Medical Applications. The main objective of CoMaMed is the development of novel optical materials which main emission is peaking within the optical window of (human) tissue. From 600 to 1300 nm tissue components like water, melanin, oxygenated and deoxygenated hemoglobine have only low absorption coefficients (Fig. 1). Fig. 2 demonstrates, that human tissue – here the hand – is transparent for the light of a red laser pointer to a certain extent. Upon using laser diodes as excitation source an absorption band is preferred to narrow lines and due to the recent advantages it should be located in the blue spectral range. Taken all this requirements into account the main focus in the project will be on activators emitting in the NIR and their sensitization by suitable co-dopants with a large absorption cross-section in the blue spectral range as well as appropriate energy levels for an efficient energy transfer to the activators.

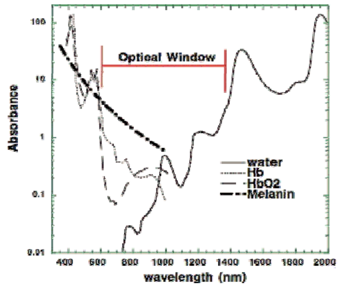

Fig. 1: Absorbance as a function of the wavelength in the visible and near infrared for components of human tissue. (www.photobiology.info/Hamblin.html (March 12th, 2009))

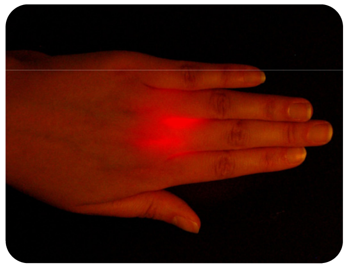

Fig. 2: Example for the weak absorbance of human tissue: Deep red light of a laser pointer is partly transmitted through a human hand

Synthesis

All samples of the system $(Ln_{1-x-y}Ce_xNd_y)_3Al_5O_{12}$ have been prepared by a combustion route employing tris(hydroxymethyl)aminomethane (TRIS). After combusting the nitrates with TRIS and sintering at elevated temperature in air or reducing atmosphere phase analysis done by x-ray powder diffraction is showing a single phase garnet type structure for all discussed samples.

Results and Discussion

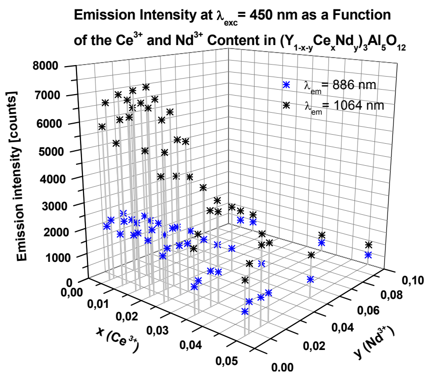

Fig. 3: The most intensive emission at 1064 nm for excitation at 450 nm has been observed for $(Y_{0,965}Nd_{0,02}Ce_{0,015})_3Al_5O_{12}$.

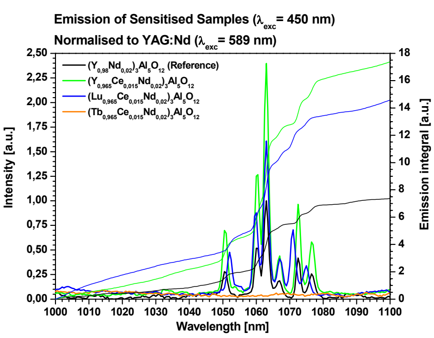

Fig. 4: Excitation of $(Y_{0,965}Nd_{0,02}Ce_{0,015})_3Al_5O_{12}$ at 450 nm yields a higher NIR intensity than excitation of $(Y_{0,98}Nd_{0,02})_3Al_5O_{12}$ at 589 nm.

Ce^{3+} is a suitable sensitizer for Nd^{3+} in garnet type hosts. Fig. 5 depicts the strong band absorption at 450 nm (which is characteristic for YAG:Ce) in both excitation spectra for the NIR emission of YAG:Ce,Nd. Excitation of YAG:Ce,Nd at 450 nm can be even more efficient than direct excitation of the activator itself in the deep red. After the absorption of the excitation energy at the sensitizing Ce^{3+} ions an energy transfer to the activator Nd^{3+} takes place and yields in an intense emission in the near infrared. Samples with varying concentrations of Ce^{3+} and Nd^{3+} have been characterised to find the optimal concentration ratio of activator to sensitizer. Fig. 3 shows that the highest emission intensities for excitation at 450 nm arise from samples with approx. 2 atom-% Ce^{3+} and 2 atom-% Nd^{3+}. The emission intensity at 1064 nm has been selected as first assessment criterion until a method for the determination of the quantum efficiencies for phosphors emitting in the near infrared will be found. Research on the effects of modifications of the host lattice, e.g. by substituting Y^{3+} by Gd^{3+}, Tb^{3+}, or Lu^{3+} as well as by replacing Al^{3+} by Ga^{3+}, or Sc^{3+} has just started. Fig. 4 shows emission spectra and integrals of modified garnets all comprising the same concentration of the sensitizer and the activator. Whereas the yttrium and lutetium garnets exhibit promising emission properties the terbium garnet does not show any NIR emission. The Nd^{3+} emission is quenched due to cross relaxation processes. The normalised emission spectra in Fig. 7 show that differences in the crystal field of the lutetium garnet compared to the yttrium garnet result in a narrowing of the Nd^{3+} multiplet at around 1064 nm.

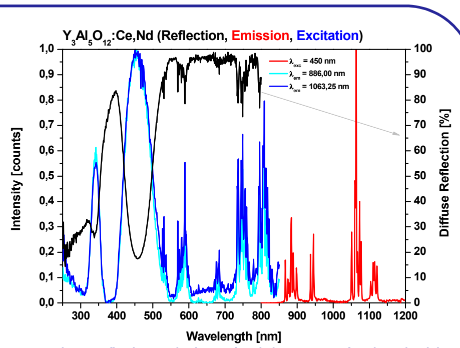

Fig. 5: Reflection, excitation and emission spectra of yttrium aluminium garnet doped with Nd^{3+} and sensitised by Ce^{3+}.

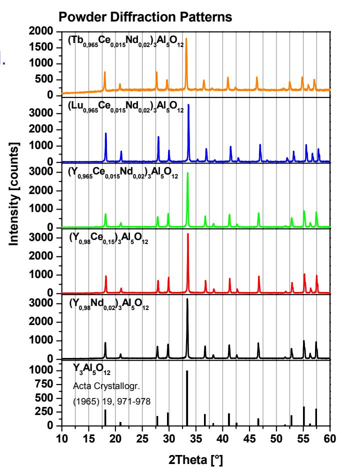

Fig. 6: All discussed samples exhibit a single phase garnet structure with high crystallinity.

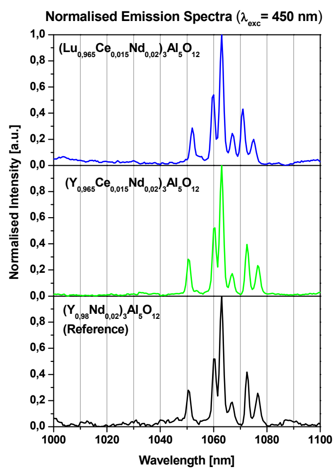

Fig. 7: Normalised emission spectra reveal a slight shift within the multiplet of the $^4F_{3/2} - {}^4I_{11/2}$ transition for the lutetium garnet.

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