On the Correlation between the Composition of Garnet Type Materials and their Photoluminescence Properties

A. Katelnikovas[a,b], D. Uhlich[a], H. Bettentrup[a], J. Plewa[a], A. Kareiva[b] and T. Jüstel[a]

Fachhochschule Münster University of Applied Sciences

[a]Department of Chemical Engineering, Münster University of Applied Sciences, Stegerwaldstr. 39, D-48565 Steinfurt, Germany

[b]Department of General and Inorganic Chemistry, Vilnius University, Naugarduko 24, LT-03225 Vilnius, Lithuania

6th Laser Ceramics Symposium, 2010 December 5-8, Münster, Germany

Introduction

Garnet is the common name of a group of cubic minerals belonging to the nesosilicates, which contain solely isolated tetrahedral $[SiO_4]^{4-}$ units. The general formula of garnets is $C_3A_2D_3O_{12}$, which, in case of natural garnets, can be written as $C_3A_2(SiO_4)_3$, where C = Fe^{2+}, Mg^{2+}, Ca^{2+} and A = Al^{3+}, Fe^{3+}, Cr^{3+} and V^{3+}. In the general formula, O denotes oxygen and C, A, D symbolize cations occupying the dodecahedral, octahedral and tetrahedral sites, respectively. Artificial garnets according to the formula $(Y_{1-x}Lu_x)_3(Al_{1-y}Ga_y)_5O_{12}$ are applied as hosts for laser gain media, scintillators, and LED phosphors.

In the present study, the luminescent properties of Nd^{3+}, Pr^{3+} and Ce^{3+} doped garnet type host lattices, namely $Lu_3(Al_{1-y}Ga_y)_5O_{12}$, $(Y_{1-x}Lu_x)_3Mg_2AlSi_2O_{12}$ and $(Y_{1-x}Lu_x)_3Al_3MgSiO_{12}$, are discussed as a function of their composition.

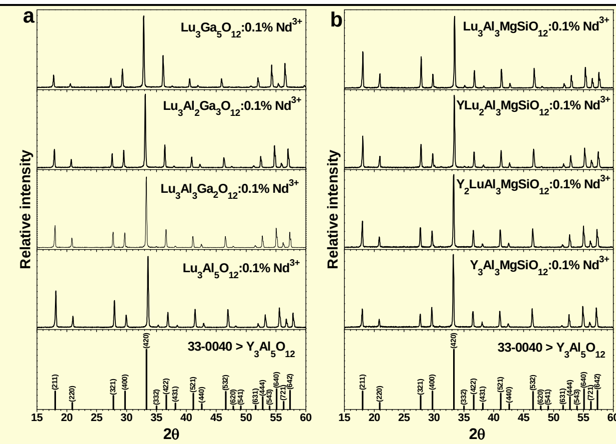

Fig.1. XRD patterns of $Lu_3(Al,Ga)_5O_{12}$ (a) and $(Y,Lu)_3Al_3MgSiO_{12}$ (b) doped with 0.1% Nd^{3+} sintered at 1600 °C.

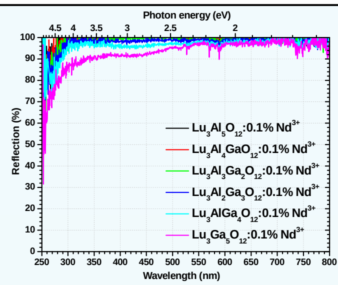

Fig. 2. Reflection spectra of $Lu_3(Al,Ga)_5O_{12}$:0.1%Nd^{3+}

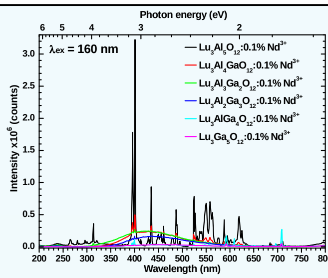

Fig. 3. Emission spectra of $Lu_3(Al,Ga)_5O_{12}$:0.1%Nd^{3+}

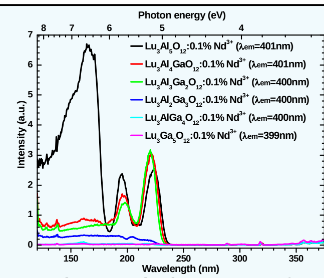

Fig. 4. Excitation spectra of $Lu_3(Al,Ga)_5O_{12}$:0.1%Nd^{3+}

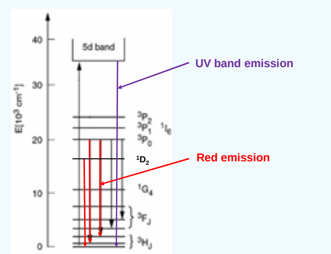

Fig. 5. Simplified Dieke Diagram of Pr^{3+} involving the lowest crystal-field component of the $[Xe]4f^15d^1$ config.

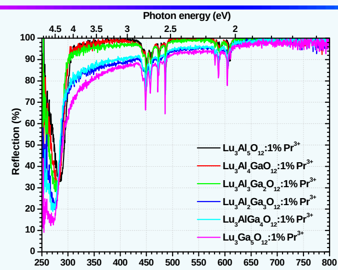

Fig. 6. Reflection spectra of $Lu_3(Al,Ga)_5O_{12}$:1%Pr^{3+}

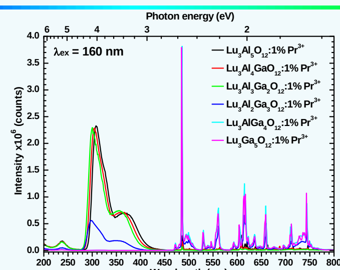

Fig. 7. Emission spectra of $Lu_3(Al,Ga)_5O_{12}$:1%Pr^{3+}

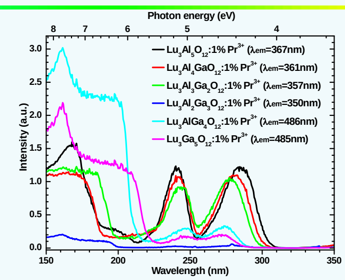

Fig. 8. Excitation spectra of $Lu_3(Al,Ga)_5O_{12}$:1%Pr^{3+}

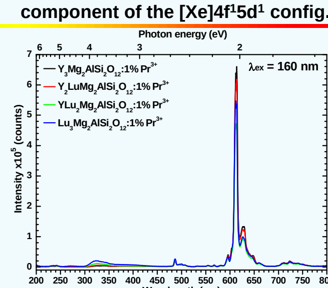

Fig. 9. Emission spectra of $(Y,Lu)_3Mg_2AlSi_2O_{12}$:1%Pr^{3+}

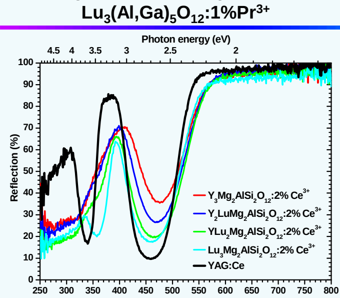

Fig. 10. Reflection spectra of $(Y,Lu)_3Mg_2AlSi_2O_{12}$:2%Ce^{3+}

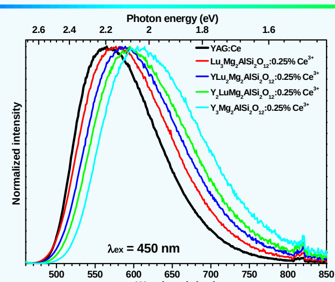

Fig. 11. Emission spectra of $(Y,Lu)_3Mg_2AlSi_2O_{12}$:0.25%Ce^{3+}

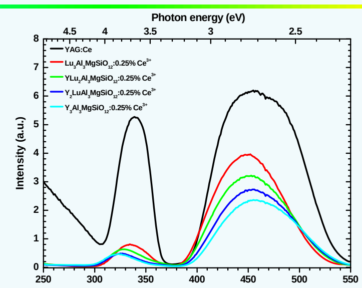

Fig. 12. Excitation spectra of $(Y,Lu)_3Al_3MgSiO_{12}$:0.25%Ce^{3+}

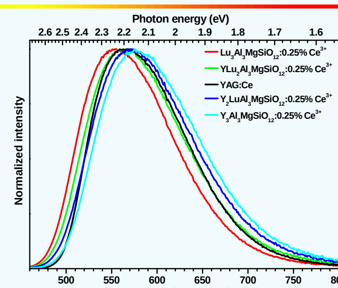

Fig. 13. Emission spectra of $(Y,Lu)_3Al_3MgSiO_{12}$:0.25%Ce^{3+}

Conclusions

In this work, we demonstrated that a modification of the LuAG host by Ga, Y, or Si and Mg results in strong changes with respect to the optical spectra of Ce^{3+}, Pr^{3+}, or Nd^{3+} located onto the dodecahedral garnet sites. Ce^{3+} emits in LuAG:Ce at around 525 nm. Any replacement of Lu by Y or Al by Mg and Si, results in a red-shift. Even a 600 nm emitting garnet is feasible by using $Y_3Mg_2AlSi_2O_{12}$ as a host. Pr^{3+} doped garnets show very peculiar spectra, due to presence of 4f-5d (UV range) and 4f-4f (visible range) transitions. The ratio of band to line emission can be easily tuned by the garnet composition. While LuAG:Pr mainly shows several 4f-5d emission bands in the UV range, in all Si/Mg comprising garnets the 4f-4f line transitions dominate the spectrum. The well known LuAG:Nd reveals intense visible emission lines upon 160 nm excitation. The replacement of Al^{3+} by Ga^{3+} results in a band gap shift from 180 to 215 nm. At the same time, the luminescence of Nd^{3+} in the visible range is completely quenched, which reveals that the ET from the host lattice excitons to the 4f-4f states of the dopant Nd^{3+} is mediated by the high lying 4f5d-levels at 195 and 220 nm.