

MATRIX INDUCED DIFFERENCES IN LUMINESCENCE PROPERTIES OF LANTHANIDE-SUBSTITUTED MIXED-METAL $L:Y_3Al_{5-x}M_xO_{12}$ ($M=In, Cr, L=Ce, Eu, Er$ AND Tb ; $0.50 < x < 2.25$) GARNETS SYNTHESIZED BY SOL-GEL METHOD

Darius Jasaitis¹, Ramunas Skaudzius¹, Aivaras Kareiva¹, Arturas Akelis¹, Thomas Justel², Arturas Katelnikovas²

¹Department of General and Inorganic Chemistry, Vilnius University, Naugarduko 24, LT-03225 Vilnius, Lithuania

²Institute for Optical Technologies, University of Applied Sciences Münster

E-mail: ramunas.skaudzius@gmail.com

Introduction

Yttrium aluminum garnet (YAG) shows exceptional chemical stability and therefore doped or substituted with Ce^{3+} , Eu^{3+} , Tb^{3+} , Cr^{3+} , Sm^{3+} , Dy^{3+} or Tm^{3+} is employed as the host material of multicolored phosphors. By selecting corresponding lanthanide ions may be produced red, green and blue (RGB) emission for use in tricolor white light [1-3].

It is known that the chemical composition of host material influences of optical properties of such phosphors considerably [4]. The matrixes of garnets could be modified by replacing different molar part of aluminium or gallium by other metals in yttrium aluminium or yttrium gallium garnets (YAG, YGG). Therefore, in the present work the sinterability, microstructural and luminescence properties evolution of lanthanide-doped and lanthanide-substituted mixed-metal $Y_3Al_{5-x}M_xO_{12}$:L ($L=Ce, Eu, Er$ or Tb ; $M=In, Cr, 0.75 \leq x \leq 2.0$) garnets powders synthesized by an aqueous sol-gel process were investigated.

Experimental

The gels were prepared using stoichiometric amounts of analytical-grade Y_2O_3 , $Al(NO_3)_3 \cdot 9H_2O$, In_2O_3 or $Cr(NO_3)_3 \cdot 9H_2O$ and $Ce(NO_3)_3$, Eu_2O_3 , Er_2O_3 and Tb_4O_7 as starting materials and 1,2-ethanediol as complexing agent. The oven dried (100 °C in air) gel powders were ground in an agate mortar and preheated for 2 h at 800 °C in air. After an intermediate grinding in an agate mortar the powders were additionally sintered for 10 h at 1000 °C.

The synthesized samples were characterized by X-ray diffraction analysis, UV-visible and emission spectra.

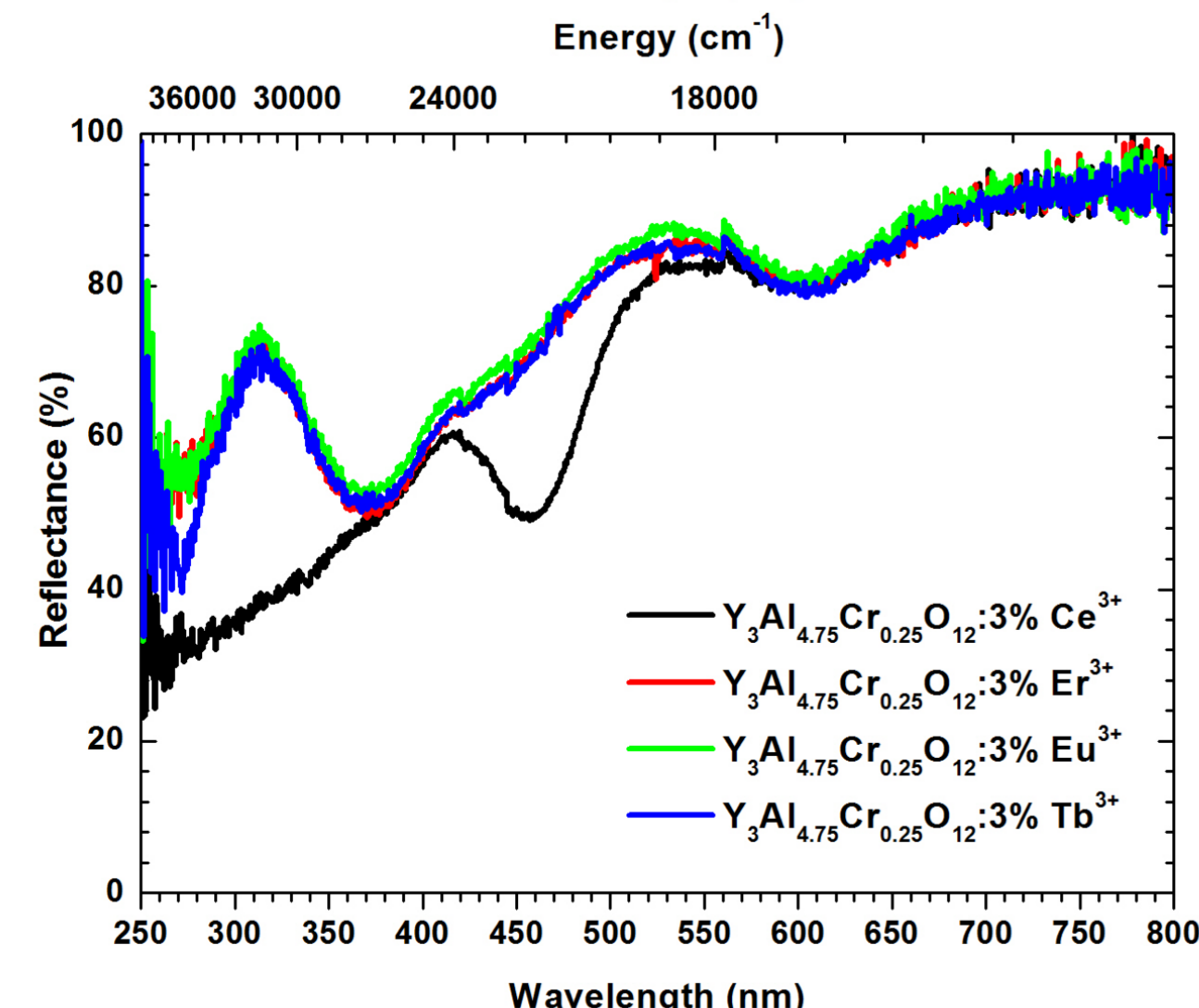
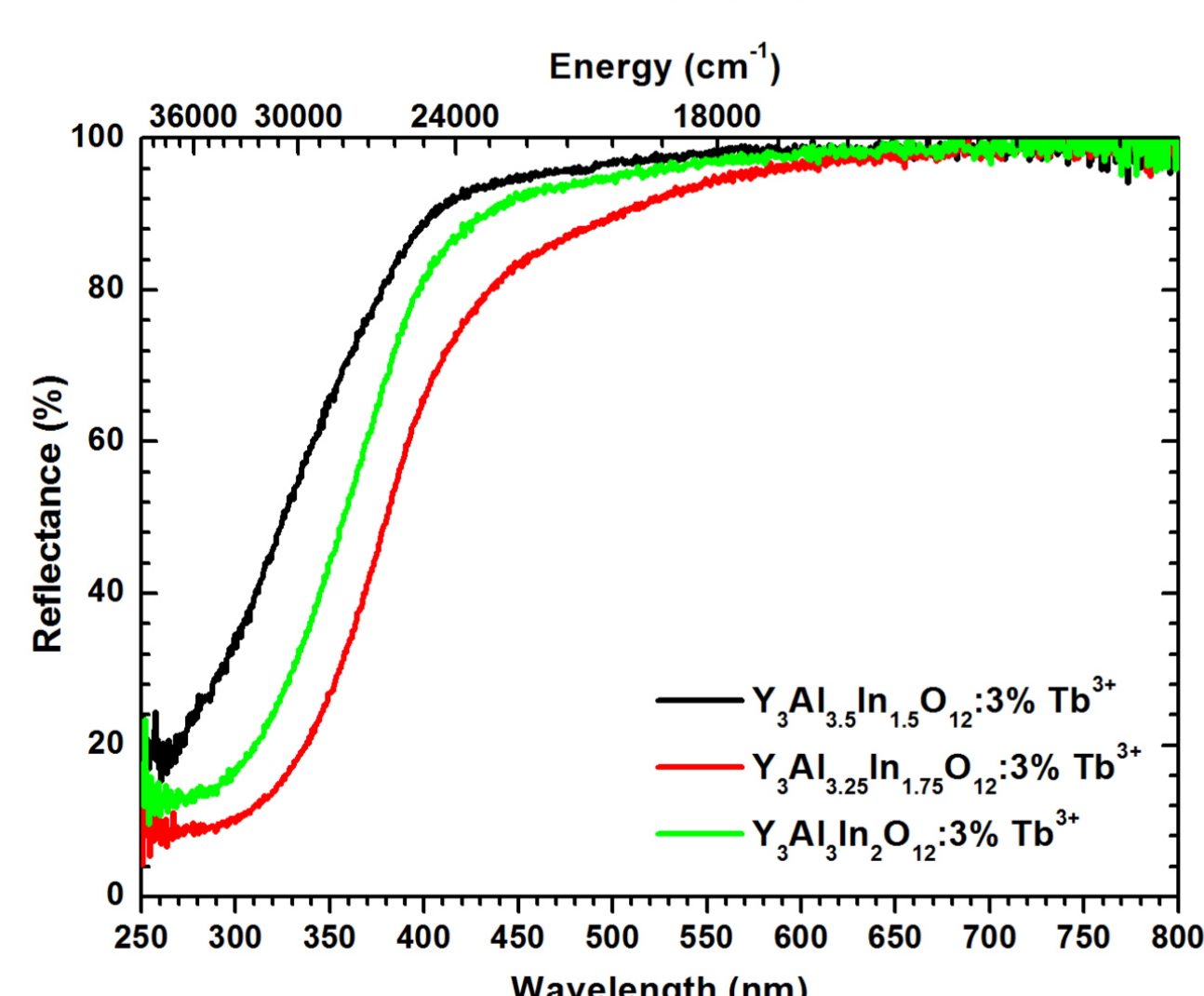
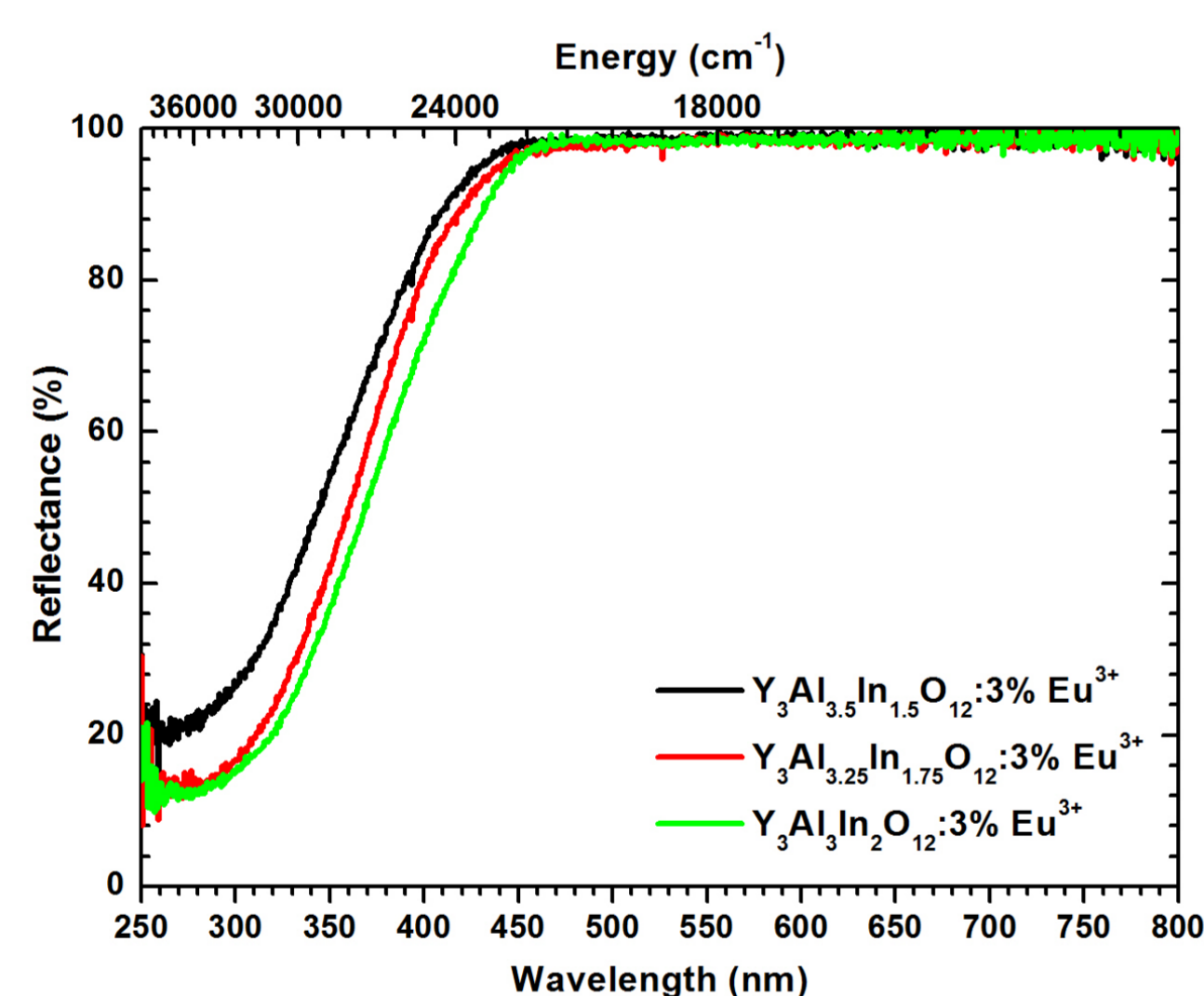
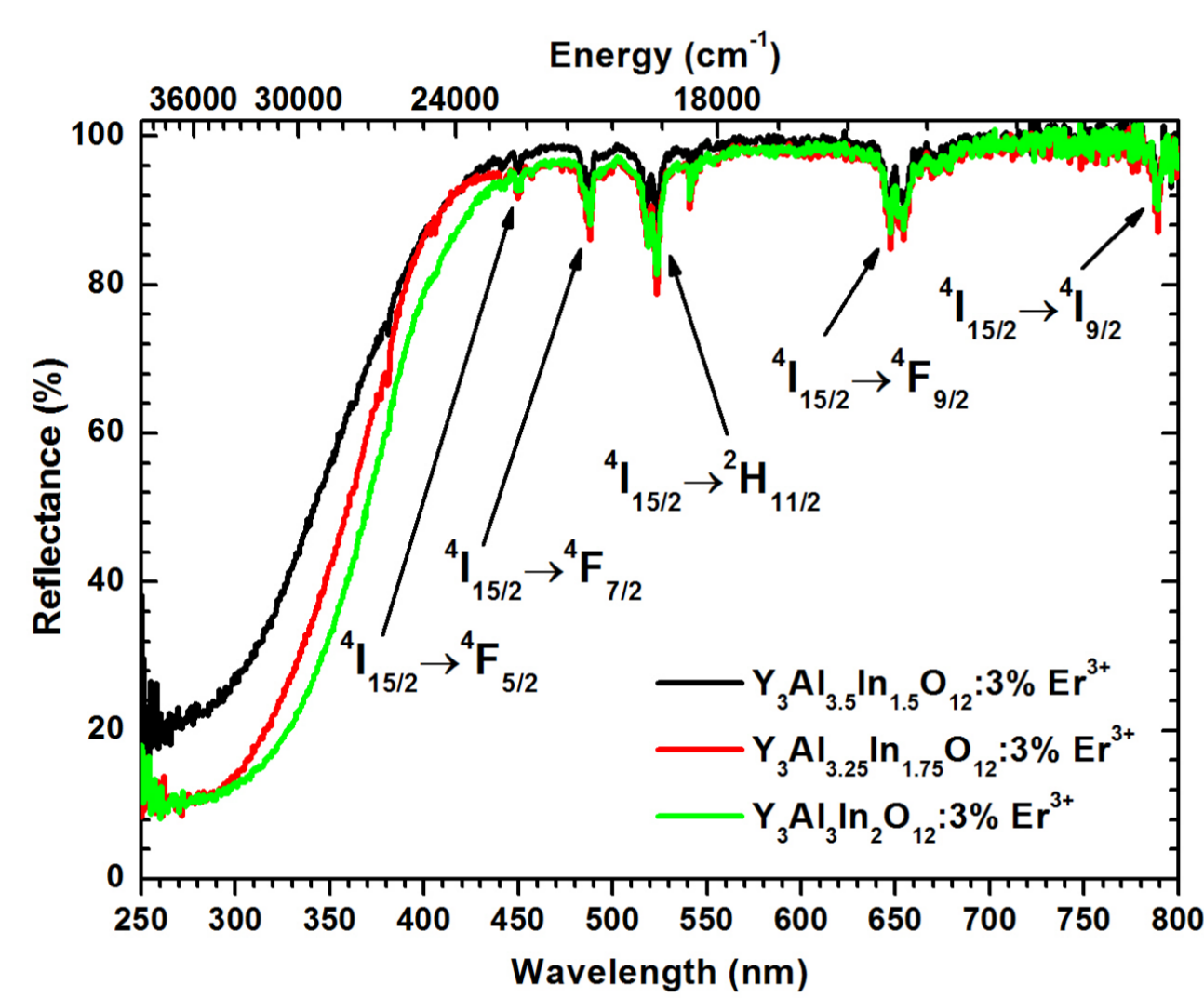
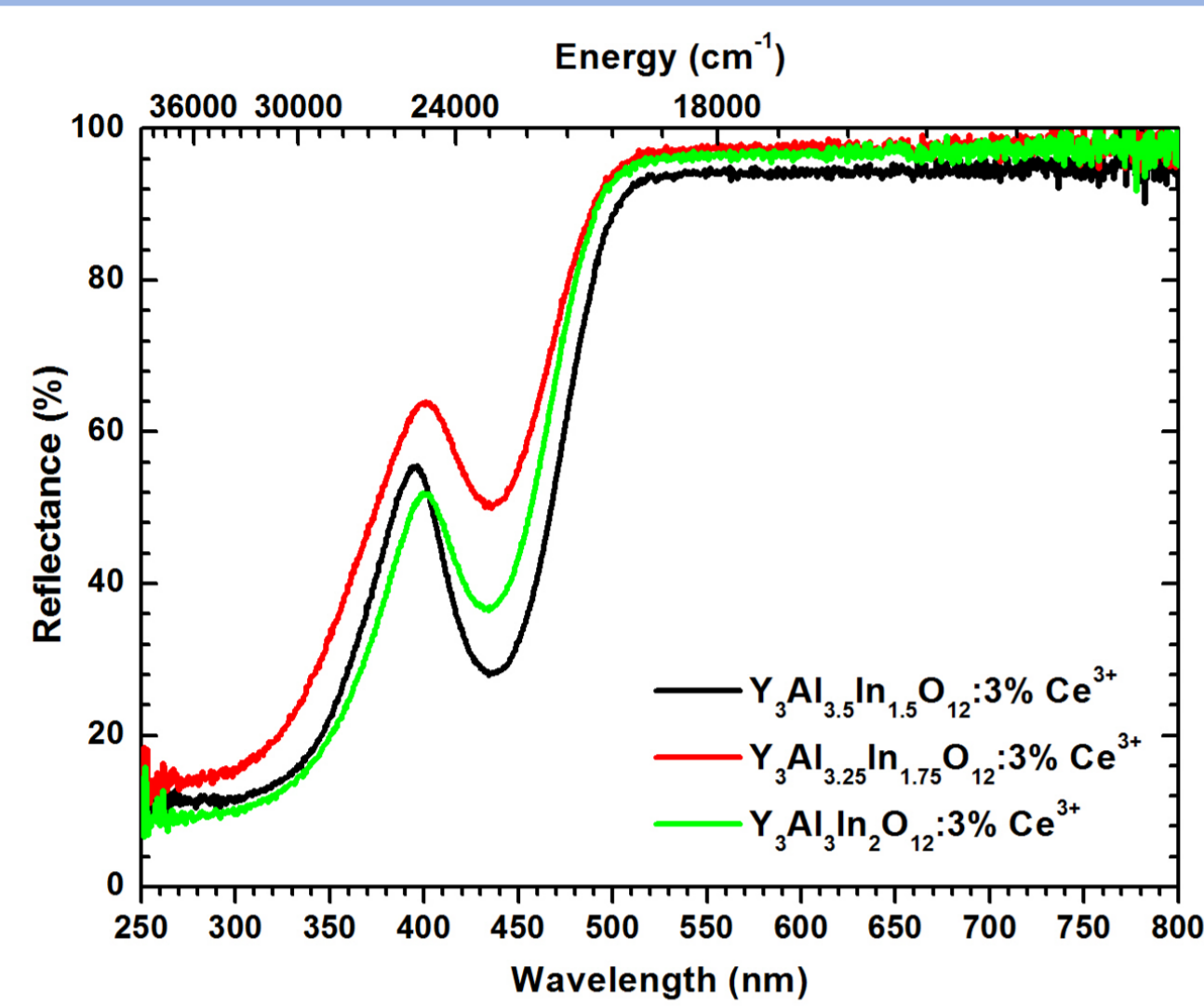
Conclusions

We have demonstrated that reflectance, excitation and luminescence spectra intensity of $L:Y_3Al_{5-x}In_xO_{12}$ ($L=Ce, Eu, Er$ or Tb) is slightly dependant on the phase purity. Though, wavelength of peaks is almost constant. When 3% yttrium is substituted by cerium in yttrium-aluminium-indium the samples have one peak with wavelength maximum at 530 nm due to $[Xe]d^5 \rightarrow [Xe]f^1$ transition. When yttrium is substituted by erbium, europium or terbium each sample contains several emission peaks. $Y_3Al_{5-x}In_xO_{12}:3\% Eu^{3+}$ samples have several peaks due to the $^5D_0 \rightarrow ^7F_j$ ($j=0, 1, 2, 3, 4$) transition between the wavelength of 580 – 720 nm. Meanwhile $Y_3Al_{5-x}In_xO_{12}:3\% Er^{3+}$ samples contains several peaks due to the $^4S_{3/2} \rightarrow ^4I_{15/2}$ transition between the wavelength of 520 – 570 nm and $^4F_{9/2} \rightarrow ^4I_{15/2}$ transition between the wavelength 650 – 680 nm. The peaks of $Y_3Al_{5-x}In_xO_{12}:3\% Tb^{3+}$ samples are between the wide wavelength of 370 – 700 nm due to the $^5D_3 \rightarrow ^7F_j$ ($j=6, 5, 4$) and $^5D_4 \rightarrow ^7F_j$ ($j=5, 4, 3, 2$) transitions. The emission spectra of $Y_3Al_{5-x}In_xO_{12}:L$ ($L=Ce, Eu, Er, Tb$) is nearly related with emission spectra of YAG:L ($L=Ce, Eu, Er, Tb$).

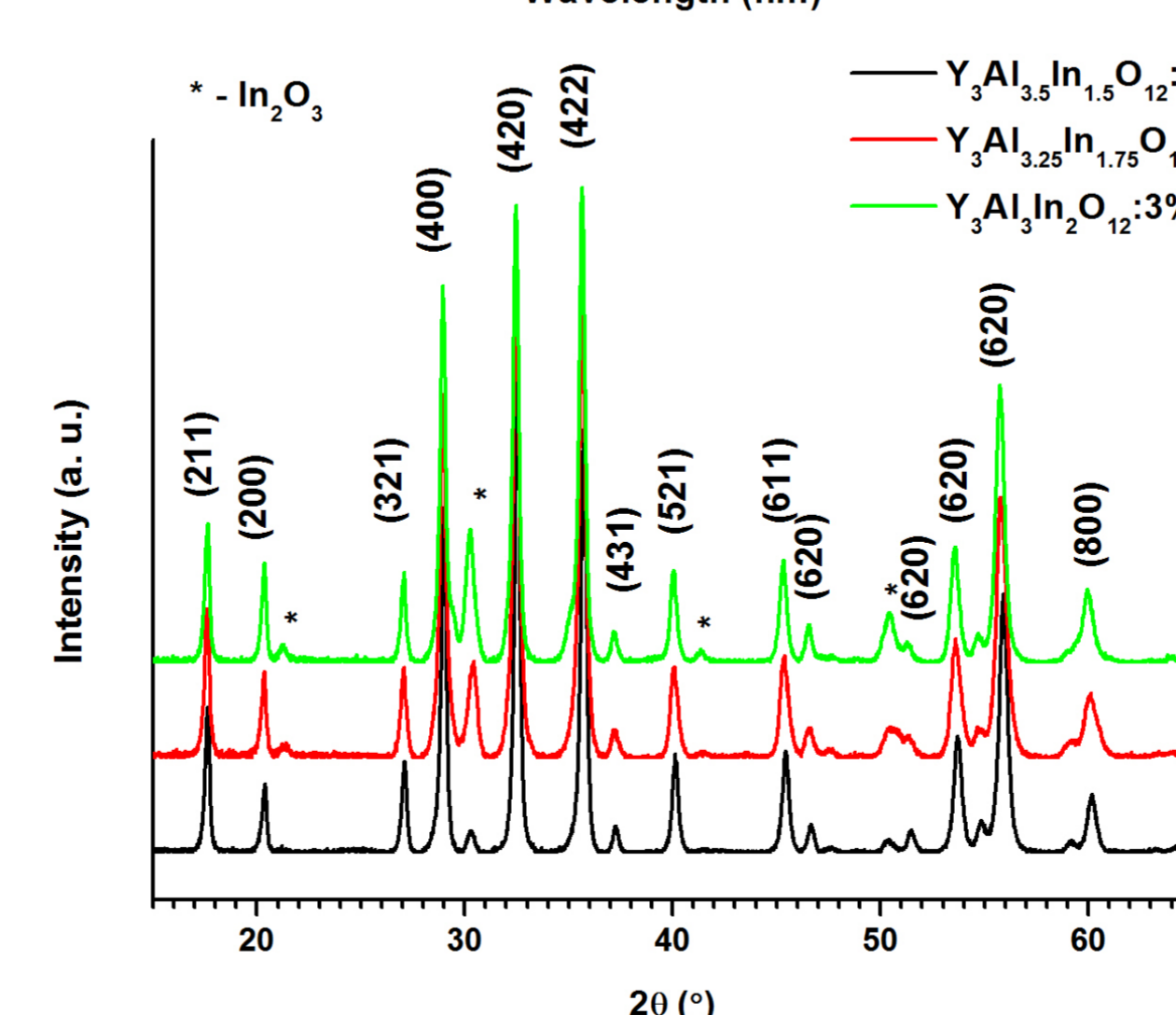
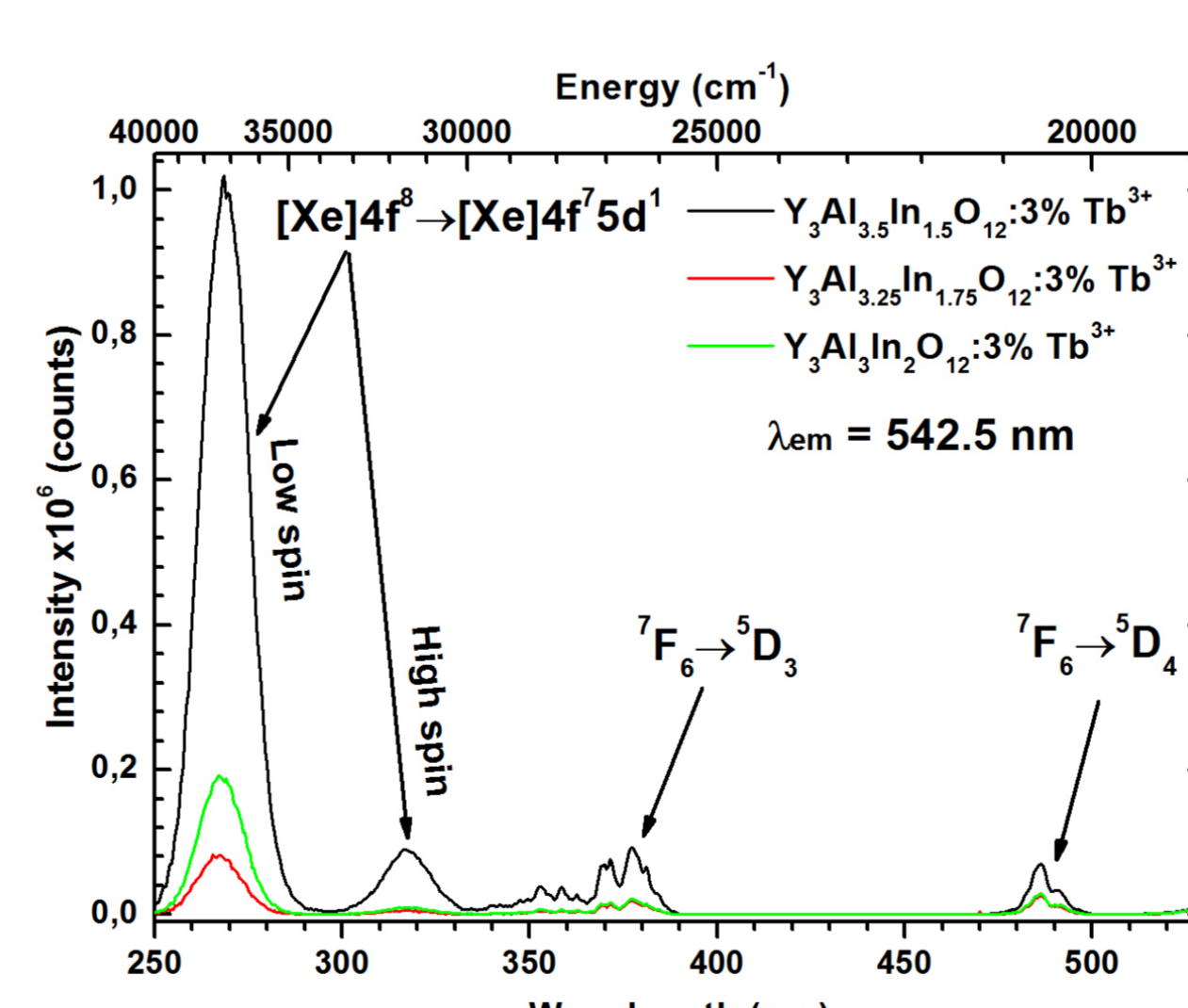
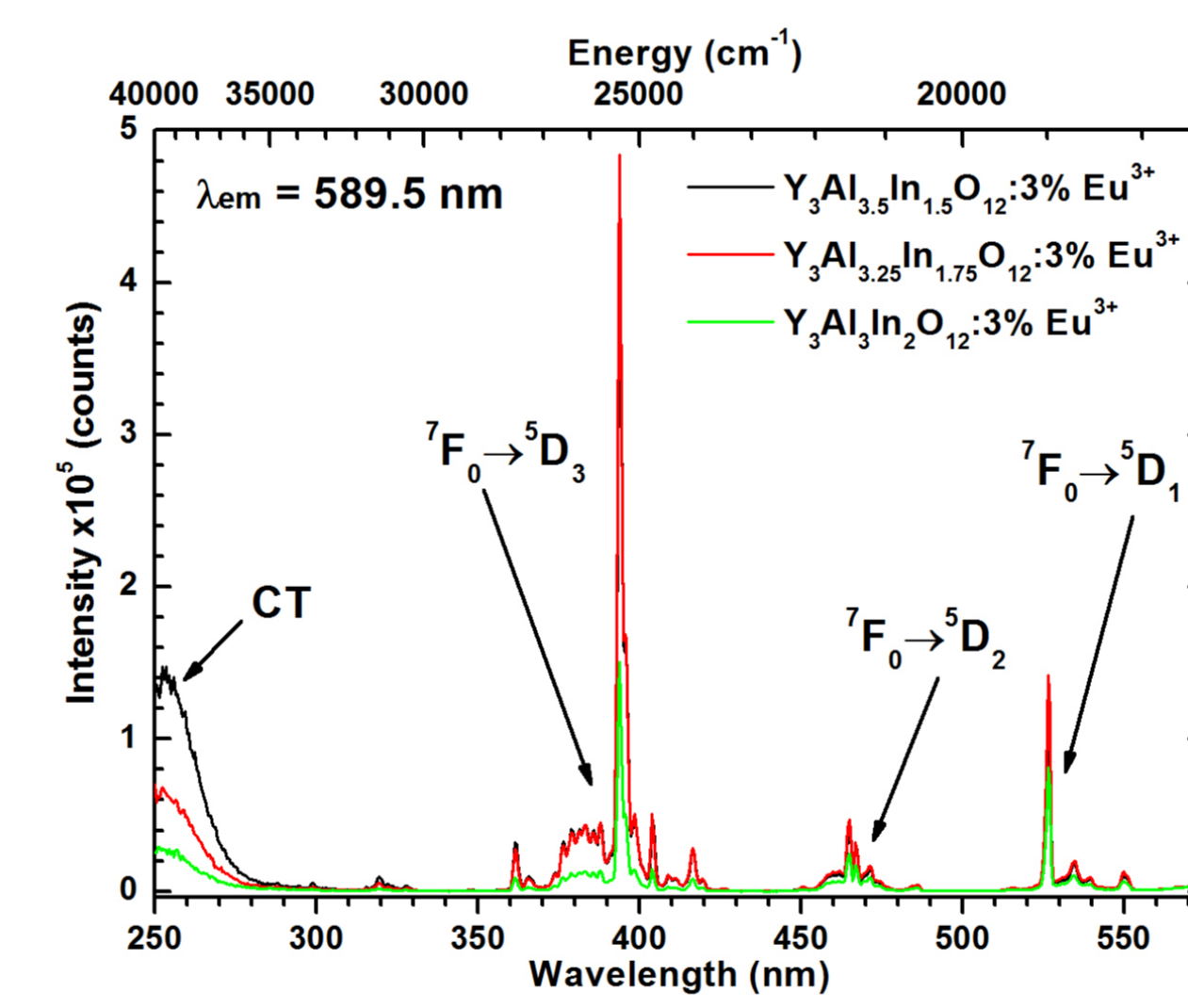
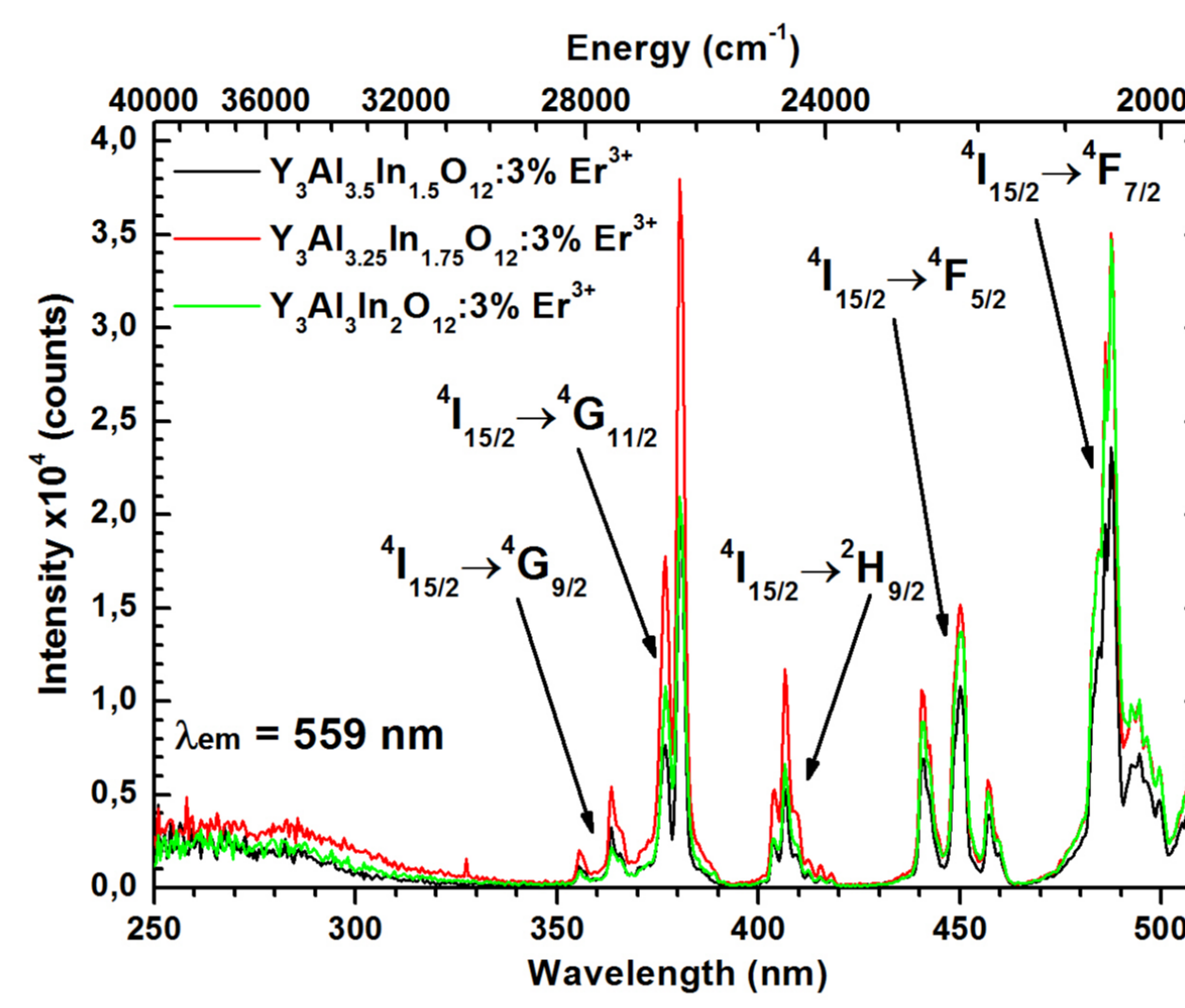
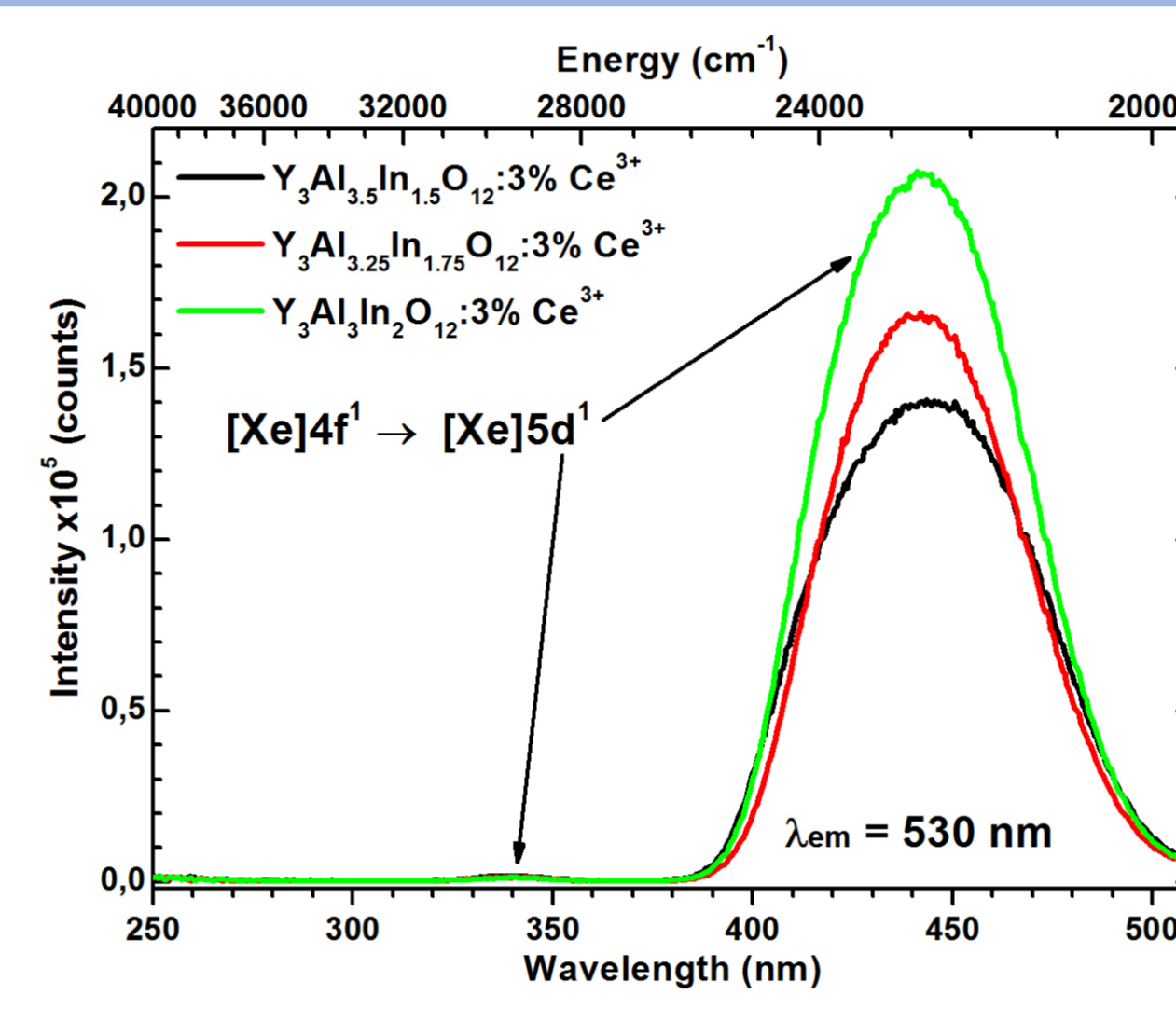
The tentative investigations showed that the modification of matrix by change aluminium with chromium dramatically influenced the luminescence properties. $Y_3Al_{4.25}Cr_{0.75}O_{12}:3\% Eu^{3+}$ is optically inactive material and does not show any emission.

Results

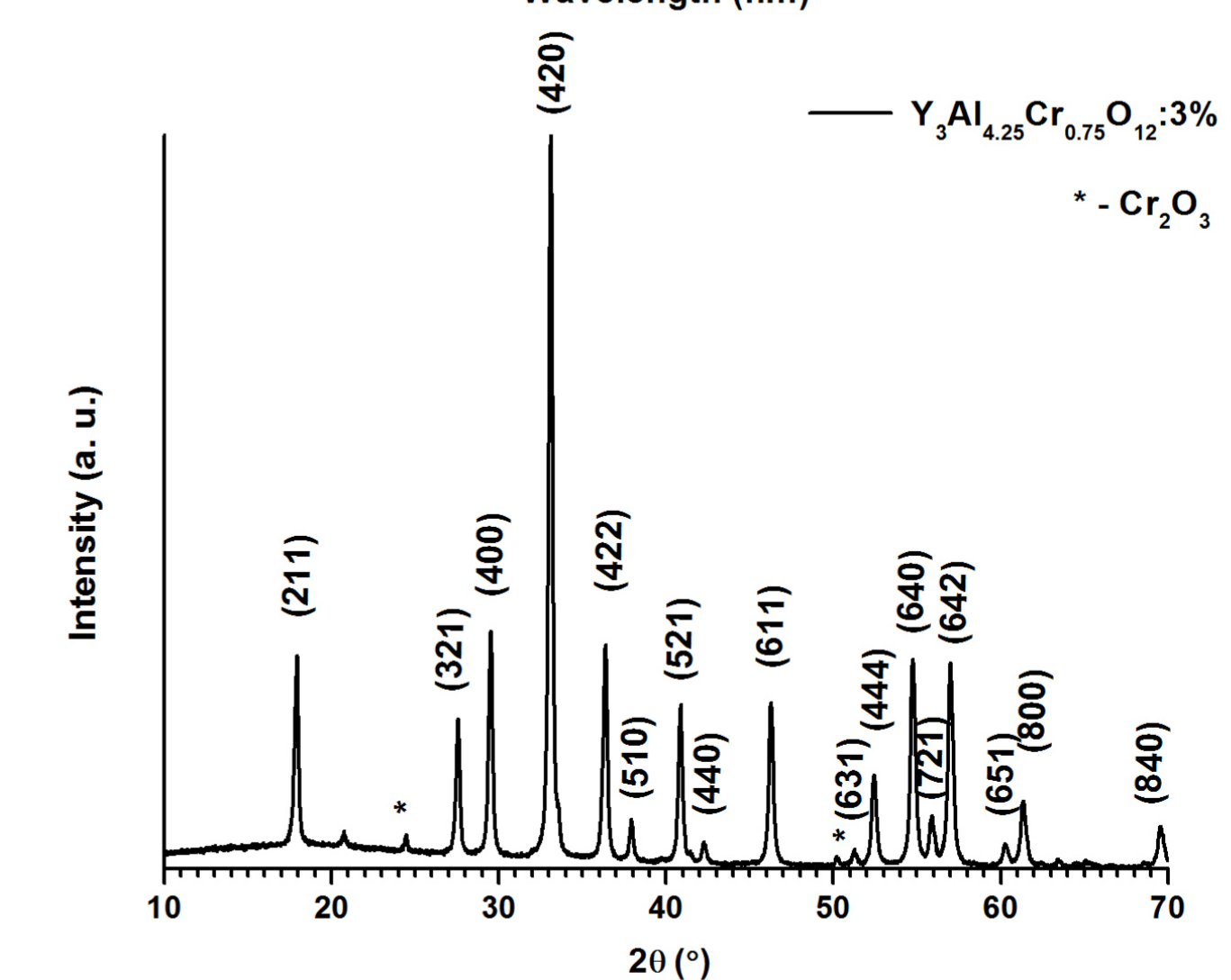
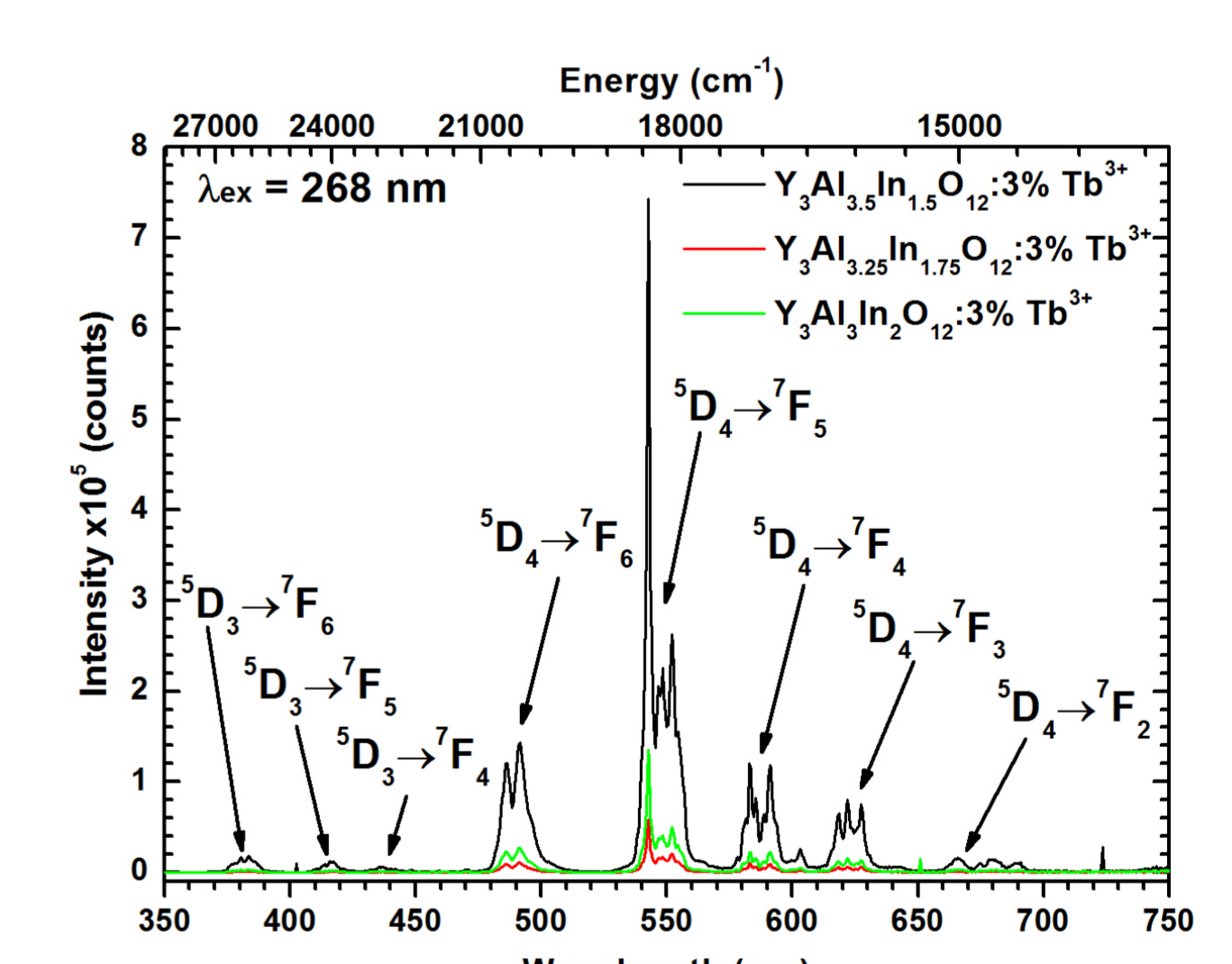
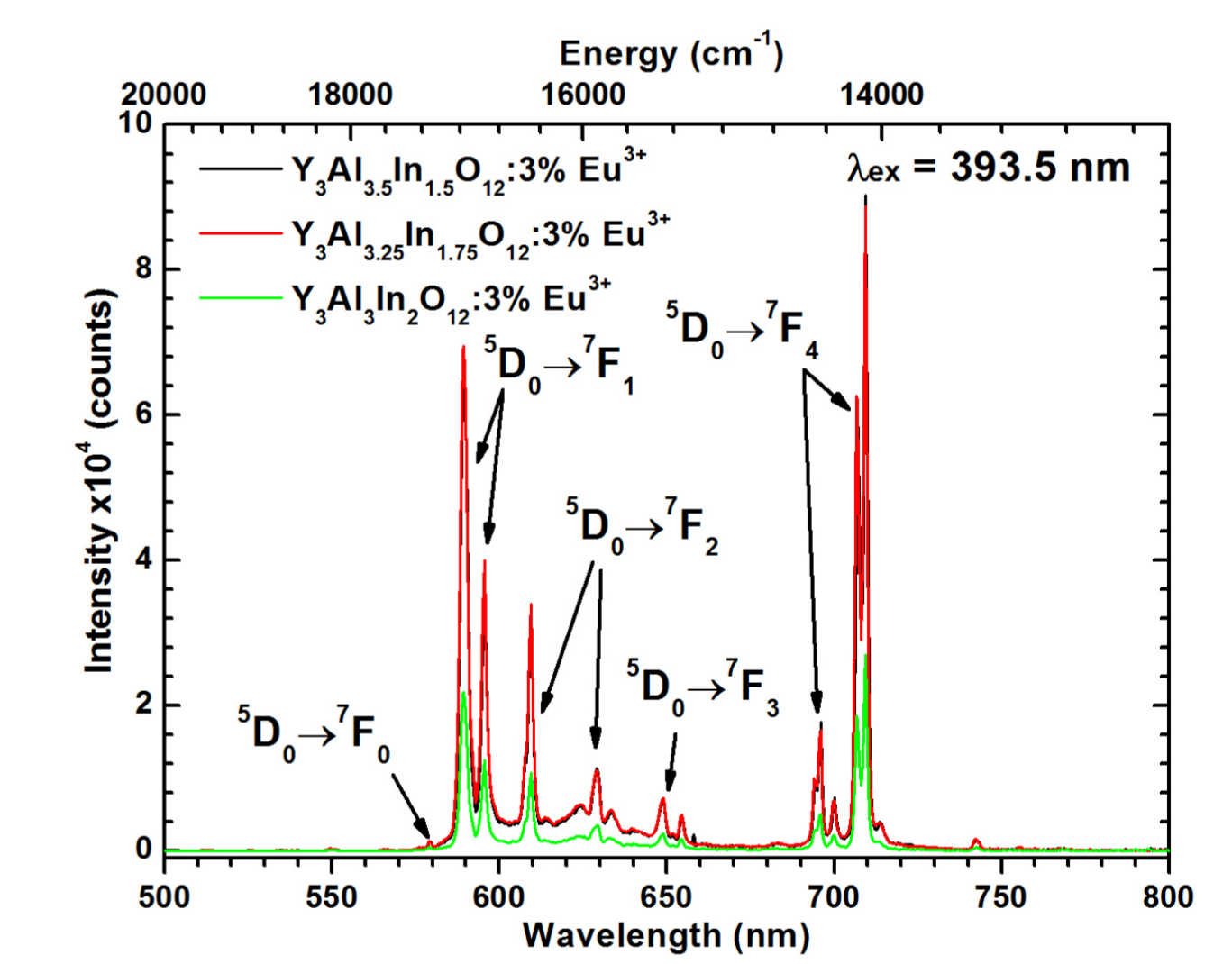
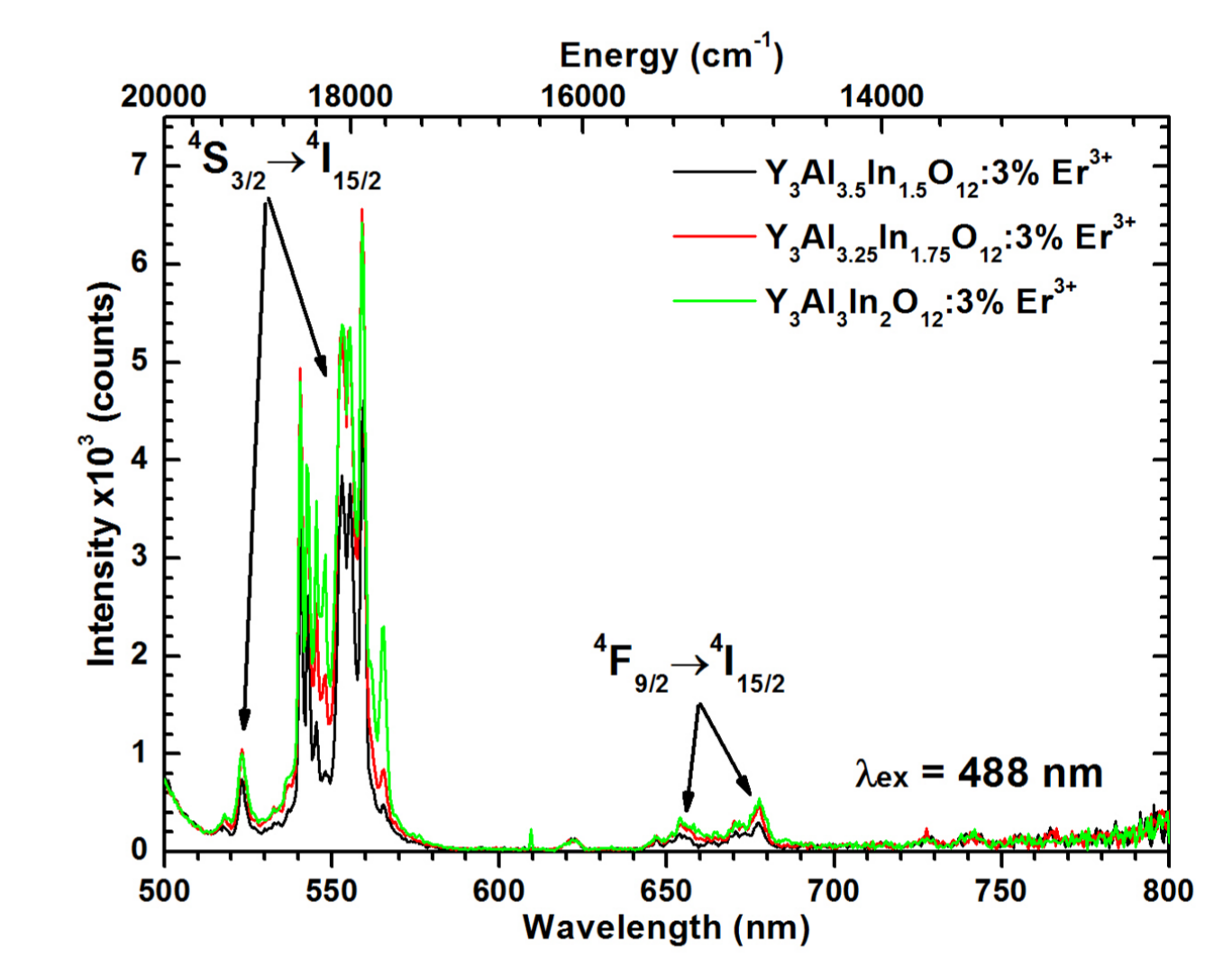
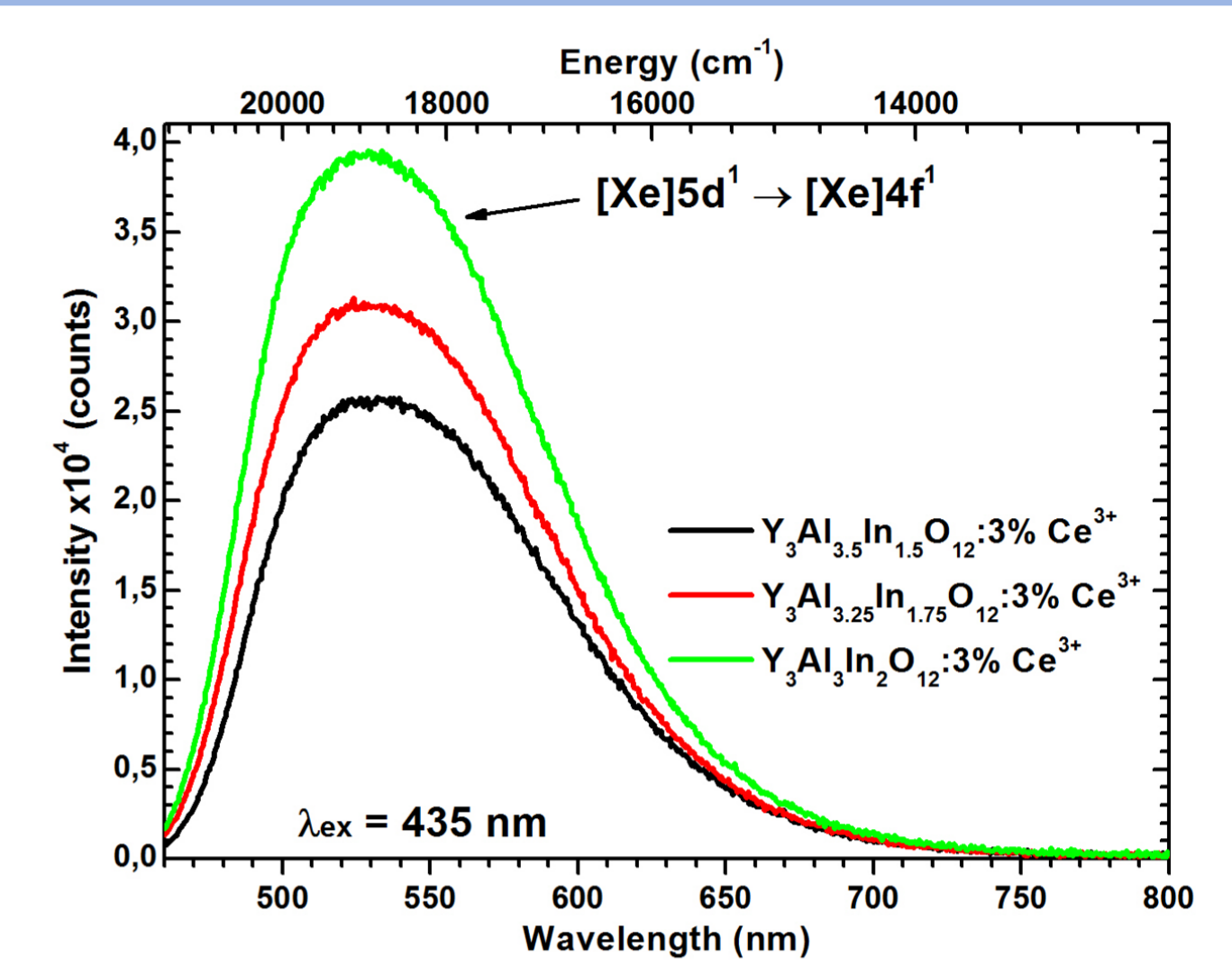
Reflectance



Excitation



Emission



References

1. W.T. Hsu, W.H. Wu and C.H. Lu, Mater. Sci. Engin., 104 (2003) p.40.
2. A. Katelnikovas, P. Vitta, P. Pobedinskas, G. Tamulaitis, A. Žukauskas, J.E. Jorgensen, A. Kareiva, J. Cryst. Growth., 304 (2007) p.361.
3. P.F.S. Pereira, M.G. Matos, L.R. Avila, E.C.O. Nassor, A. Cestari, K.J. Ciuffi, P.S. Calefi and E.J. Nassar, J. Lumin., 130 (2010) p.488.
4. A. Katelnikovas, H. Bettentrop, D. Dutzak, A. Kareiva, T. Jüstel, J. Lumin., (2011) 10.1016.