Growth of mixed garnet and perovskite crystals

SUCCESS workshop “Lyon 2012”
December 12, 2012, Lyon

(presented by A.G. Petrosyan)
Institute for Physical Research
Academy of Sciences of Armenia

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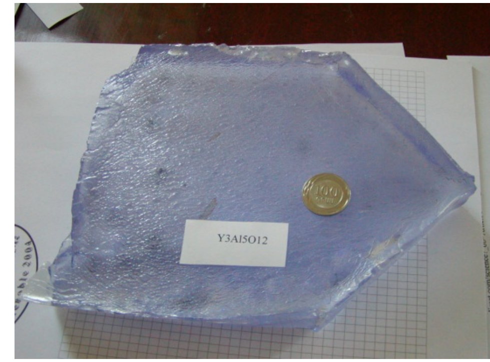


Garnets: $RE_3Al_5O_{12}$ (RE = Lu, Yb, Tm, Er, Ho, Dy, Tb, Y), $Lu_3(Al_{2-x}Sc_x)Al_3O_{12}$, $Y_3Sc_2Al_3O_{12}$, $Gd_3Sc_2Al_3O_{12}$, $Y_3Ga_5O_{12}$, $Lu_3Ga_5O_{12}$, $Gd_3Ga_5O_{12}$

Perovskites: $LaAlO_3$, $LaLuO_3$, $YAlO_3$, $LuAlO_3$, $(Y,Lu)AlO_3$, $TmAlO_3$, $YbAlO_3$, $LaGaO_3$

Orthorombic Niobates: $Ca(NbO_3)_2$

Tungstates: $CaWO_4$, $SrWO_4$

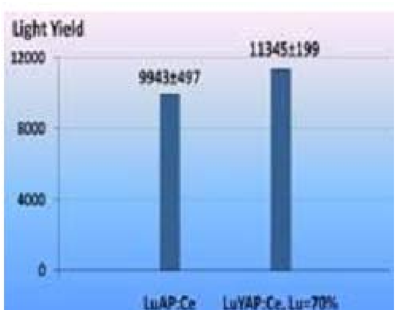
Molybdates: $NaLa(MoO_4)_2$

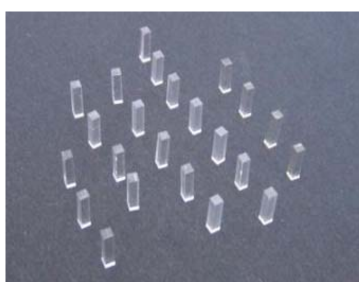
Ca Aluminates: $CaYAlO_4$, $CaNdAlO_4$

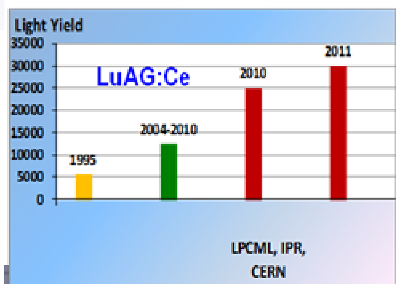
Fluorides: LiF, $LiYF_4$, LaF_3

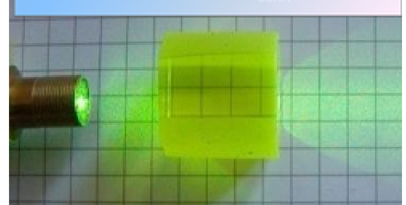
Activator ions: Ce, Pr, Nd, Sm, Eu, Ho, Er, Tm, Yb

Vertical Bridgman Technique



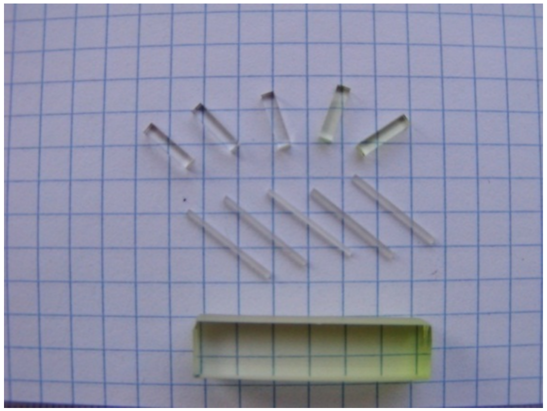




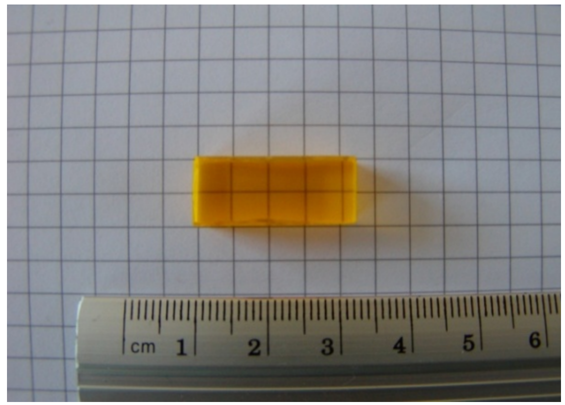


Vertical Bridgman technique

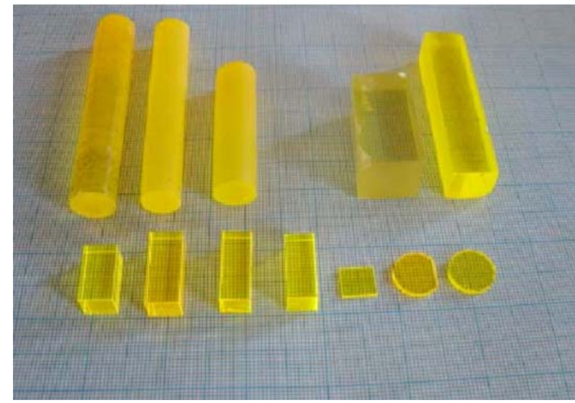
- **Boule size : diameter 10 – 24 mm; length 50 - 100 mm; Mo tubes; Ar/H2**



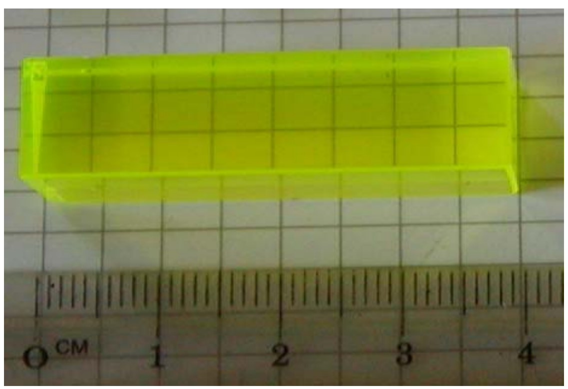
Lu3Al5O12:Pr



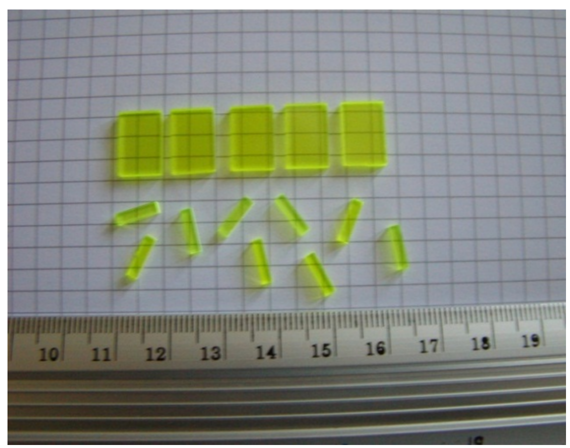
Gd3Sc2Al3O12:Ce



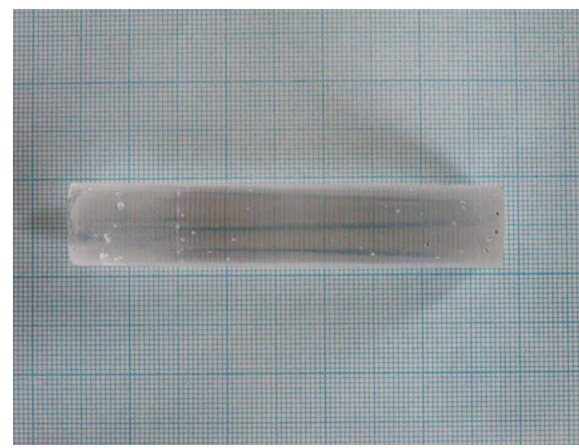
Y3Al5O12:Ce



Lu3Al5O12:Ce



Lu3Al5O12:Ce



LuAlO3:Ce


SPECORD

Solid solution systems

$YAlO_3$-$LuAlO_3$:Ce – A.Belsky et al, 2001

$(Lu,Gd)_3(Al,Ga)_5O_{12}$:Ce – K.Kamada, et al, 2011

LuAG-LuGG:Ce – M.Fasoli et al, 2011

$(Lu,Y)_3(Ga,Al)_5O_{12}$:Ce – K.Kamada, et al, 2011

LSO-YSO:Ce - J.Chen, et al, 2005

LSO-GSO:Ce - G.Loutts, et al, 1997; O.Sidletskiy, et al, 2010.

Solid solution garnets under current studies with single- or multi-site substitutions

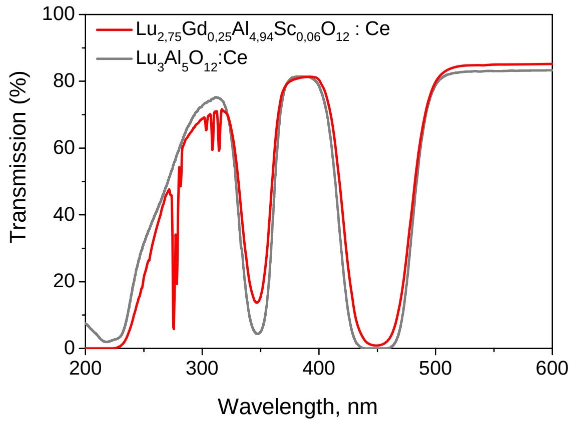
$\{Lu,Y\}_3 [Al]_2 (Al)_3 O_{12}$: Ce *30000 ph/MeV (Lu80/Y20) (CERN)*

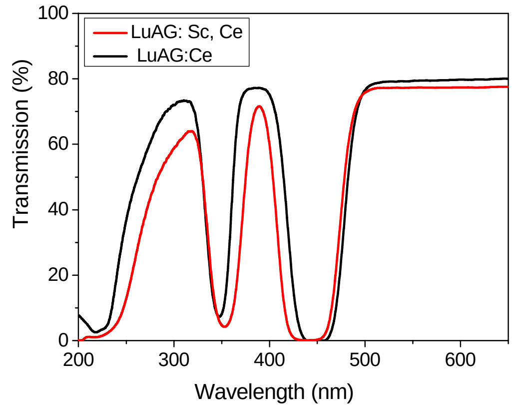
$\{Gd\}_3 [Al,Sc]_2 (Al)_3 O_{12}$: Ce *LY = 15000 ph/MeV (CERN)*

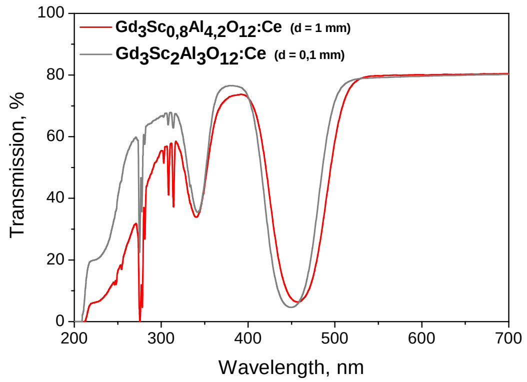
$\{Lu,Gd\}_3 [Al,Sc]_2 (Al)_3 O_{12}$: Ce

$\{Lu,Y\}_3 [Al,Sc]_2 (Al)_3 O_{12}$: Ce

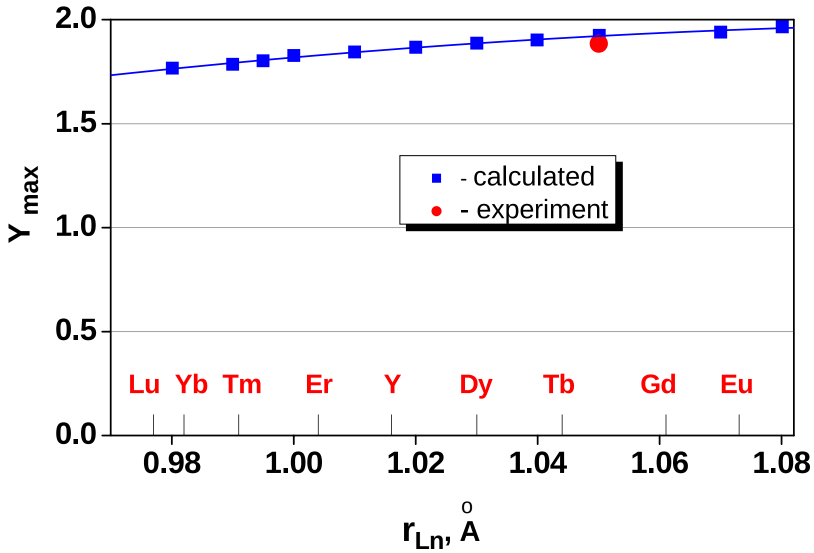
$\{Lu,Sc\}_3 [Al,Sc]_2 (Al)_3 O_{12}$: Pr or Ce







Site occupation in $Ln_3Al_5O_{12}$: Sc



$\{Ln_{3-x}Sc_x\}[Sc_2](Al_3)O_{12}$: x is increasing from 0.15 to 0.33 for Ln= Eu to Dy

C.D. Brandle, R.L. Barns, "Crystal stoichiometry and growth of rare-earth garnets containing scandium", J. Crystal Growth 20 (1973) 1-5.

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$\{Ln_{3-x}Sc_x\}[Al_{2-y}Sc_y](Al_3)O_{12}$

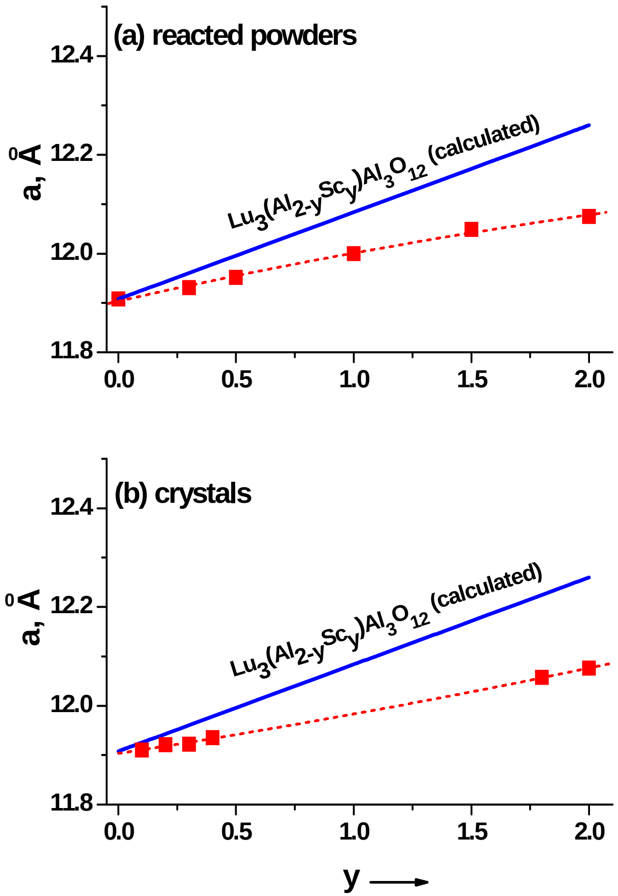
y=f(rLn); y max = 1.983 r(Ln) – 0.171

y max = 1.89 (Ln=Gd);

y max = 1.75 (calculated for Ln=Lu)

Figure from G.M.Kuzmicheva, A.A.Sattarova, "Calculation of limits for isomorphous substitutions" Vestnik MITXT, 4 (2009) 95

Lattice parameters and phase composition of samples with starting composition Lu3Al2-yScyAl3O12 and prepared by different means



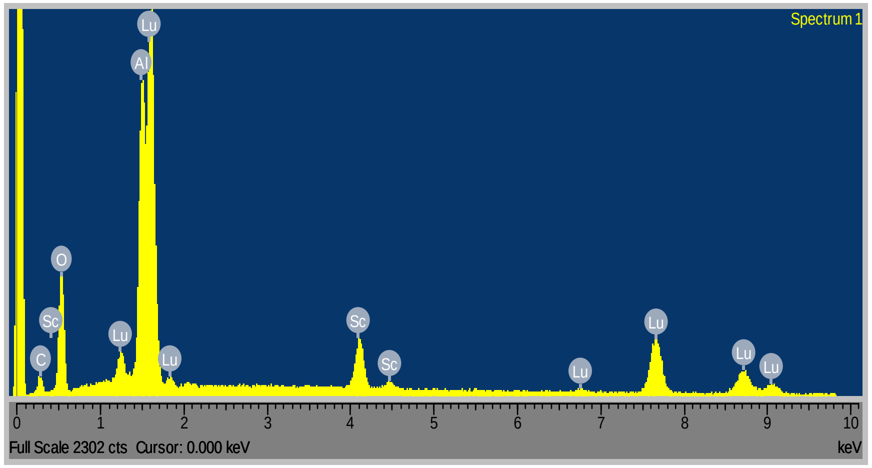
y	Reacted powders		Crystals	
	a_o (Å)	Phases	a_o (Å)	Phases / quality
0.1			11.910	G/ clear
0.2			11.921	G/ clear
0.3	11.931	G	11.922	G/ clear
0.4			11.935	G+ss(traces)/ clear
0.5	11.952	G		
1	12.001	G+ ss traces		
1.5	12.049	G + ss		
1.8			12.06÷12.08	G + ss/ opaque
2	12.075	G + ss	12.07÷12.08	G + ss/ opaque

G – garnet, ss - (Lu,Sc)2O3 solid solution

Site occupation and solubility limit of Sc in $Lu_3Al_5O_{12}$

Composition, site distribution and lattice parameters of $\{Lu_{3-x}Sc_x\}[Al_{2-y}Sc_y](Al_3)O_{12}$

Melt		Crystal composition (at%)				Calculated parameters			$a_{o\ meas}$ (Å)
x	y	Lu	Sc	Al	O	x	y	a_o (Å)	
0	0.3	14.80	0.91	24.29	60.00	0.040	0.142	11.930	11.922
0.3	1.3	13.58	6.03	20.39	60.00	0.284	0.922	12.048	12.039



X-ray microanalysis

$$\{C_{3-x}C'_x\}[A_{2-y}A'_y]D_3O_{12}$$

$$a = b_1 + b_2r^{VIII} + b_3r^{VI} + b_4r^{IV} + b_5r^{VIII}r^{VI} + b_6r^{VIII}r^{IV}$$ (B.Strocka, et al, 1978)

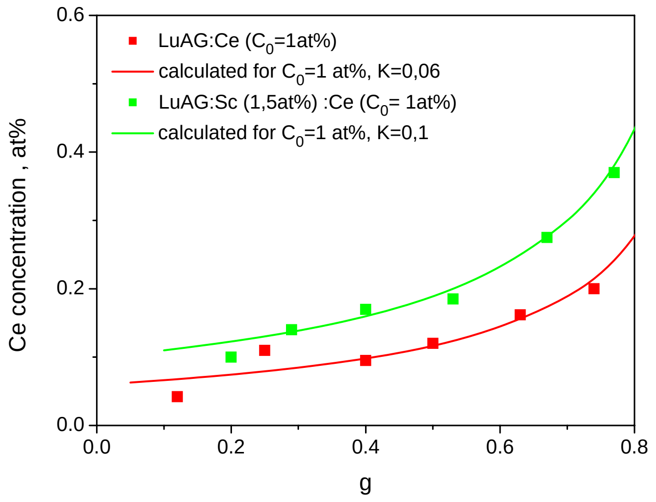
$$r_{eff}^{VIII} = r_{Lu}^{VIII} + x(r_{Sc}^{VIII} - r_{Lu}^{VIII})/3;$$

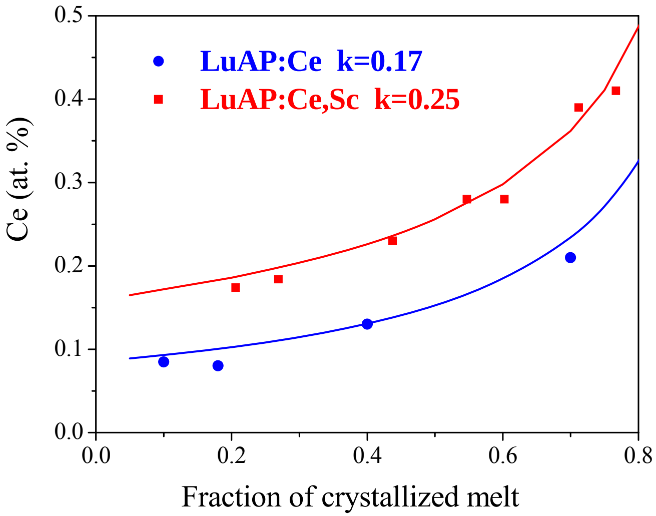
$$r_{eff}^{VI} = r_{Al}^{VI} + y(r_{Sc}^{VI} - r_{Al}^{VI})/2$$

$$a = 11.90781 - 0.08226\,x + 0.17593\,y + 0.00336xy$$

Site occupation and solubility limit of Sc in Lu3Al5O12, A.G.Petrosyan, K.L.Ovanesyan, G.O.Shirinyan, R.V.Sargsyan, C.Dujardin, C.Pedrini, J. Crystal Growth 338 (2012) 143-146

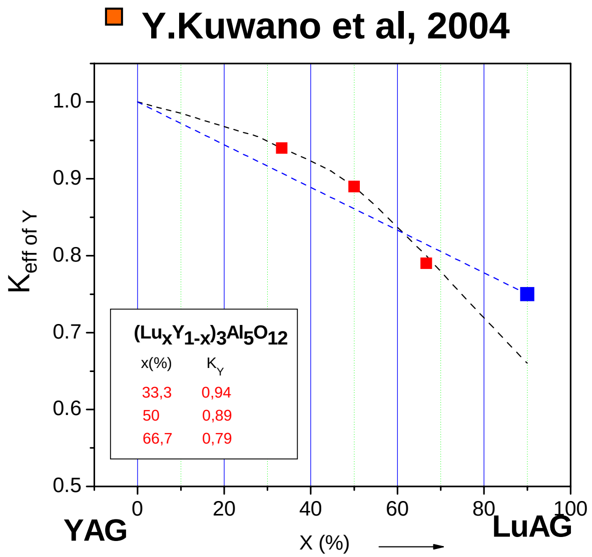
Ce distribution

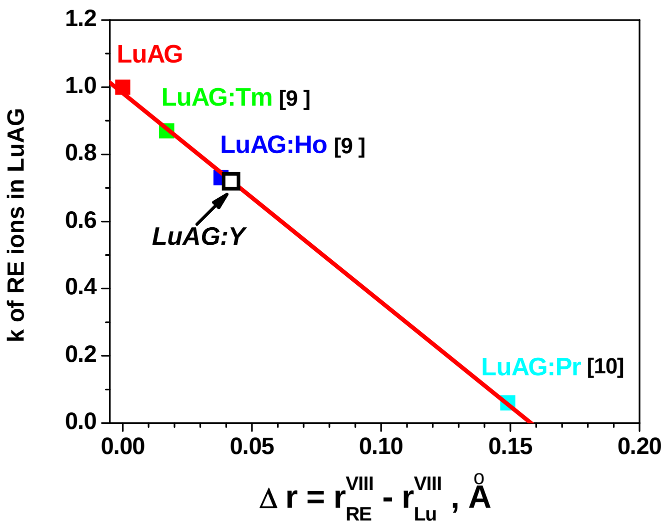


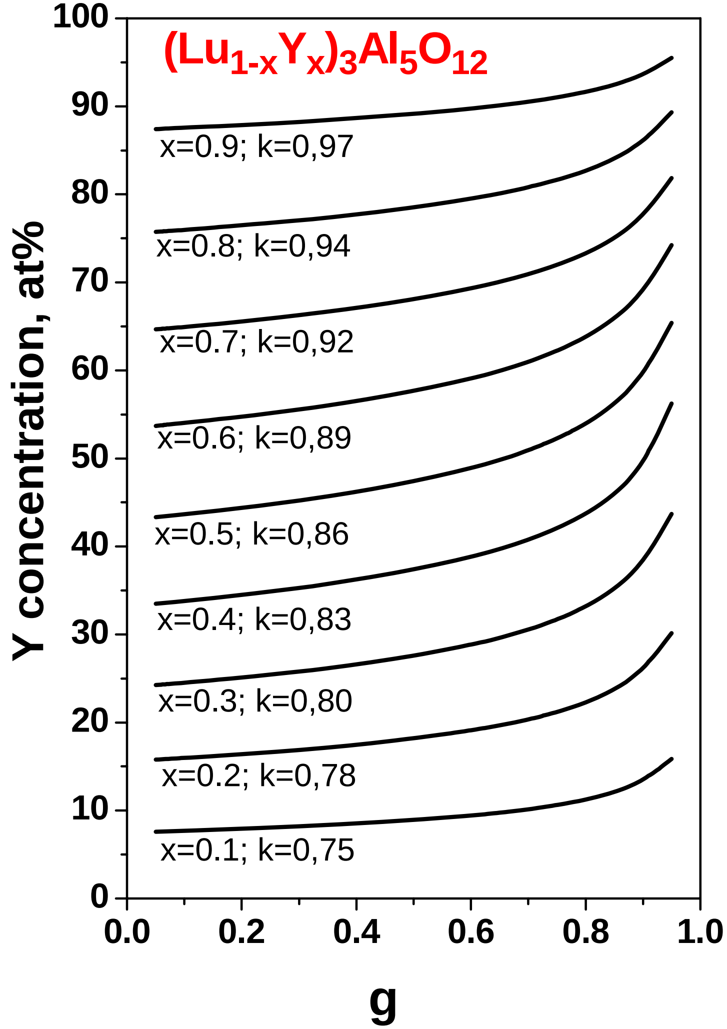


When introduced to LuAG, Sc goes to both 8- and 6-fold sites of the lattice; in 8-fold sites it leads to size compensation (Ce is larger than Lu, Sc is smaller than Lu); in 6–fold sites it leads to enlargement of the unit cell volume.
Both favor an increase of the Ce distribution coefficient.
Size compensation in Lu sites is seen also in LuAP:Ce,Sc.

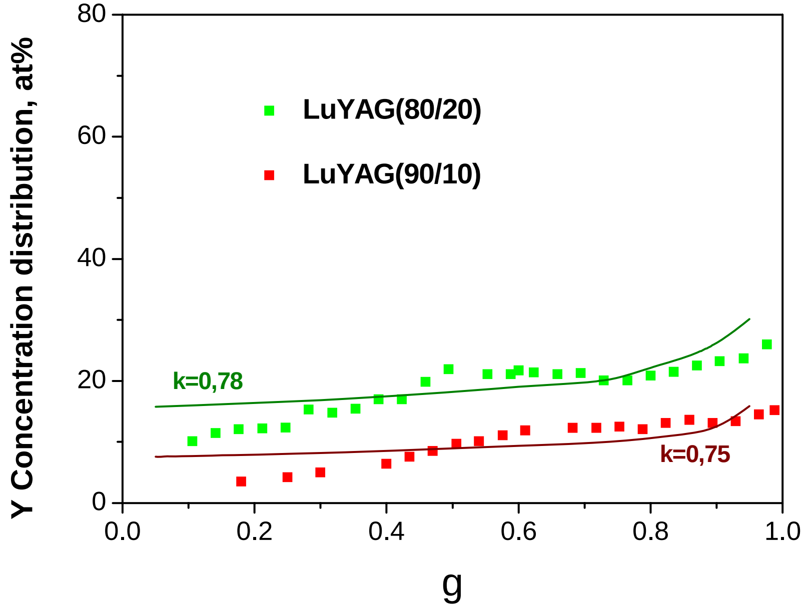
Distribution of components in (Lu,Y)AG







Distribution of components in (Lu,Y)AG in Bridgman geometry



Growth and studies of mixed (Lu,Y)3Al5O12:Ce scintillator crystals
K.L.Ovanesyan, G.R.Badalyan, A.V.Yeganyan, A.G.Petrosyan, A.Belsky, C.Dujardin, E.Auffray, P.Lecoq, K.Pauwells, N.Di Vara, poster at Laser Physics 2012 Conference, October 2012

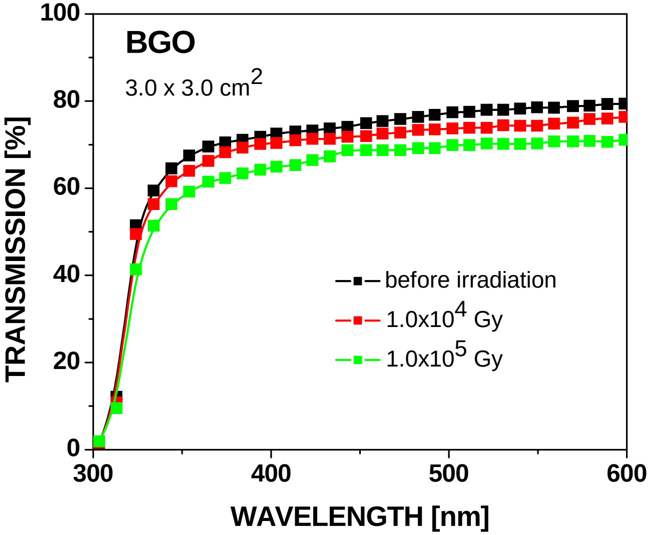
Radiation stability

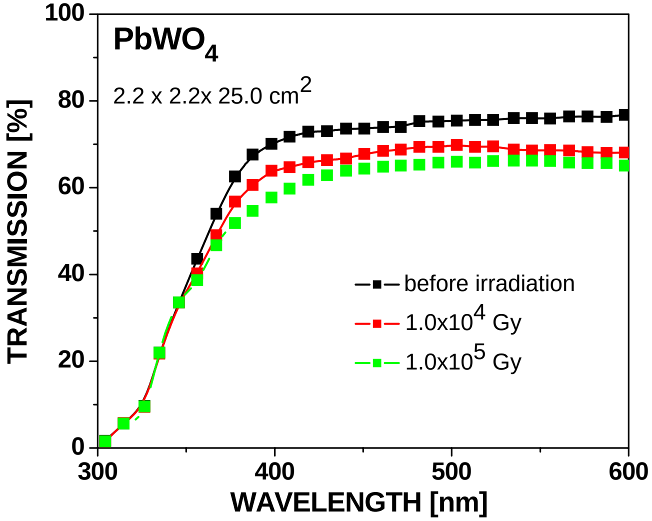
crystal	radiation dose, rad
PbWO4	>2.10^6
CsI(Tl)	10^7
BGO	10^5 - 10^6
YAG	10^{14}
Gd2SiO5:Ce	>10^9
Lu2SiO5:Ce	10^8
YAlO3:Ce	≥10^5

Radiation doses, causing noticeable decrease of the scintillation light yield
(data of different authors; ref. M.Globus et al, Inorganic Scintillators for Modern and Traditional Applications, Kharkiv, 2005; T.Hase et al, 1990)

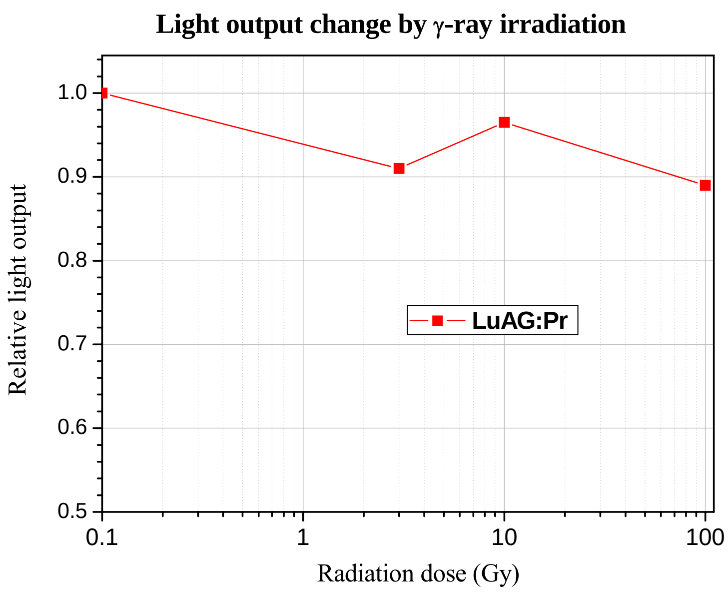
Radiation damage in scintillation crystals is due to accumulation of defects produced by irradiation. Their concentration is limited by recombination processes of produced defects.

BGO and PbWO4



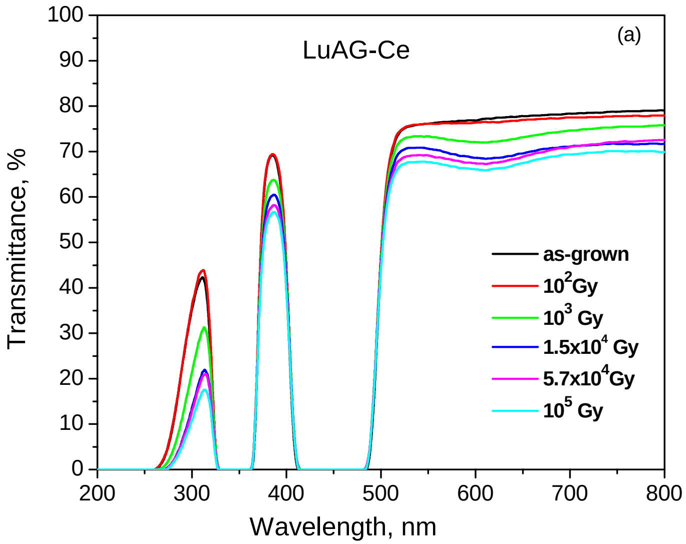


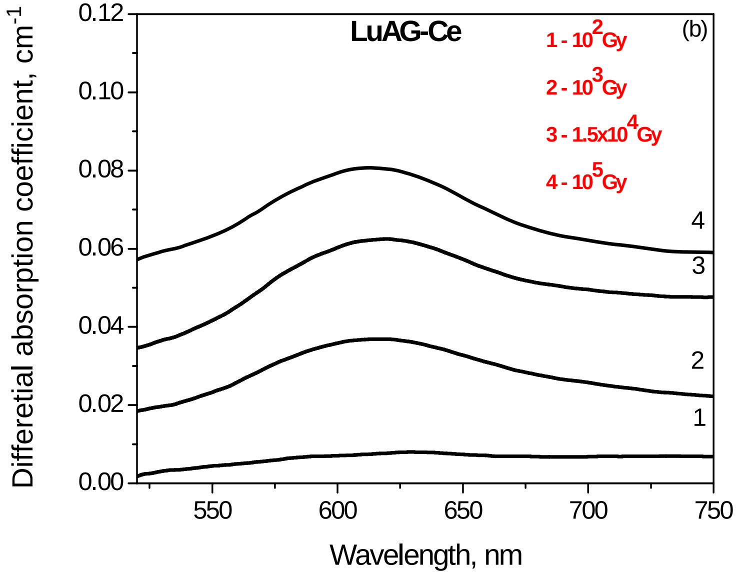
T.Iwashita, K.Miyabayashi, "Radiation Hardness Test of Pr:LuAG and BSO scintillators", Nuclear Science Symposium Conference Record (NSS/MIC) IEEE, 2010, 278-279.

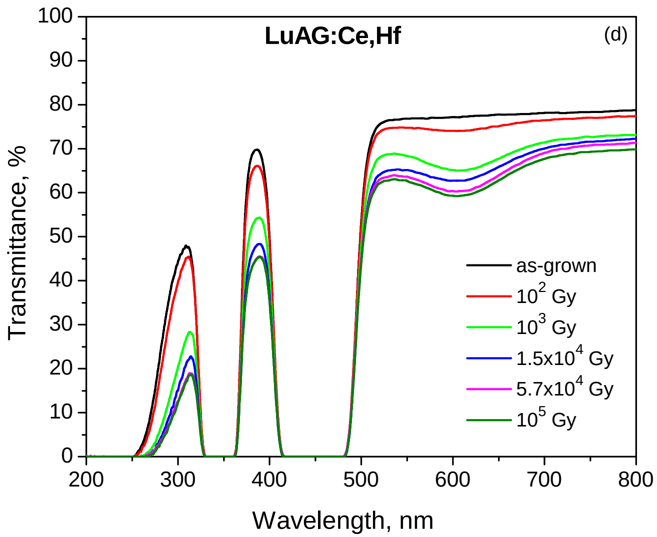


The light yield degradation of Pr:LuAG single crystal exposed to 100 Gy from ^{60}Co was (-11±3)%

Radiation induced centers in LuAG

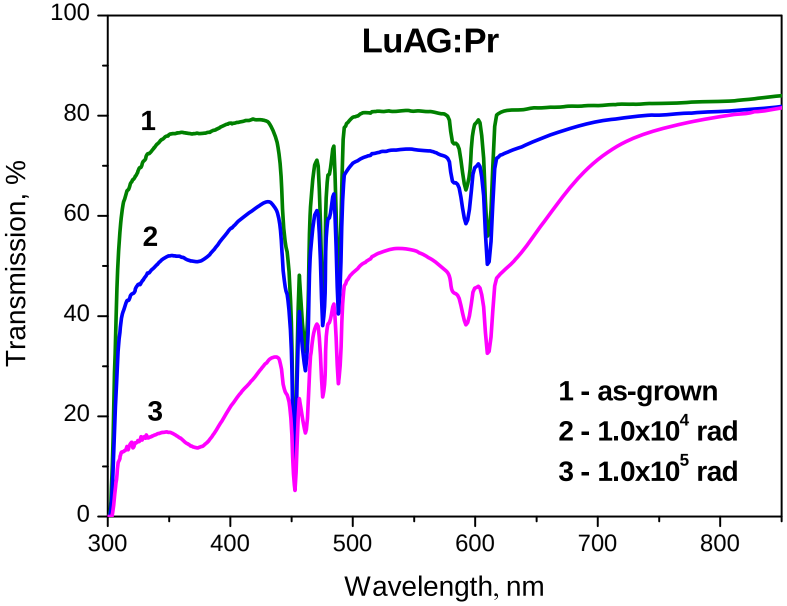


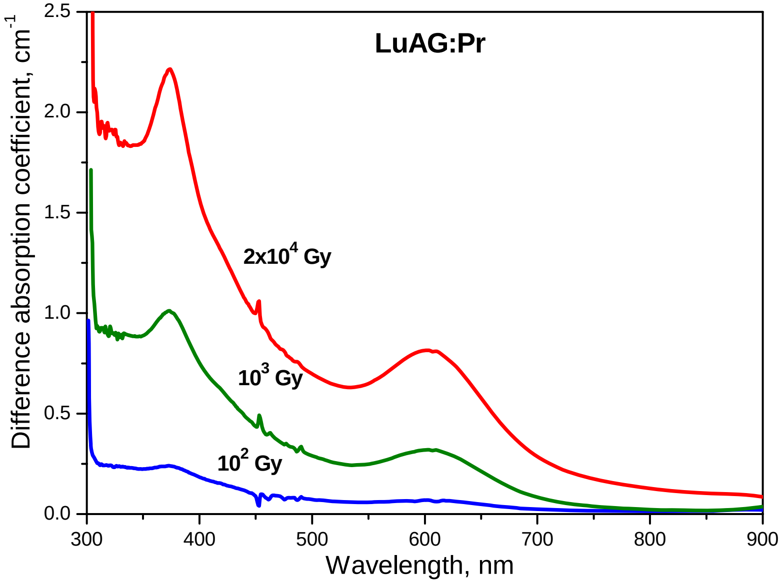




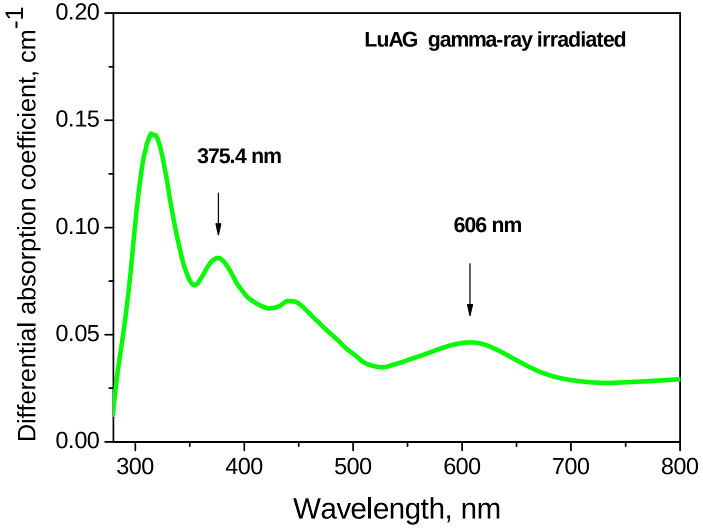
Derdzyan et al, J. Crystal Growth, 361 (2012) 212

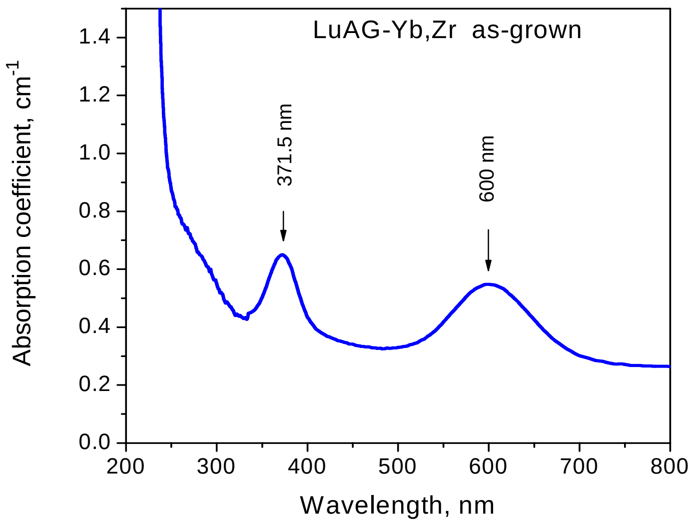
Radiation induced centers in LuAG

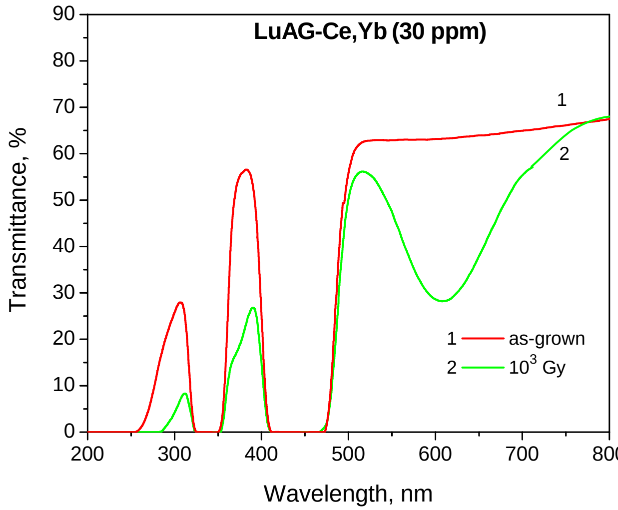




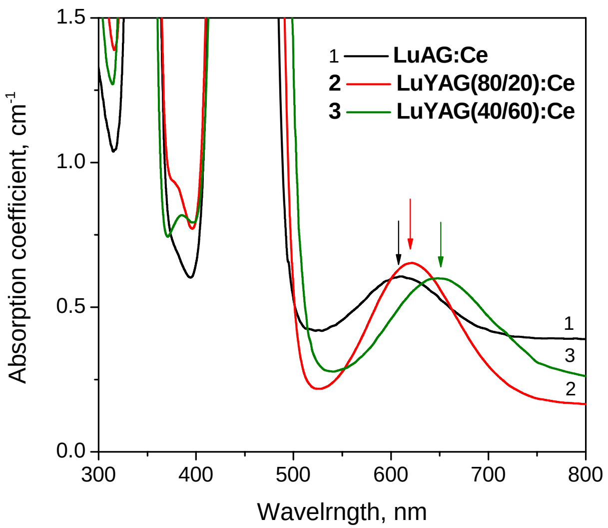
Radiation induced centers in LuAG



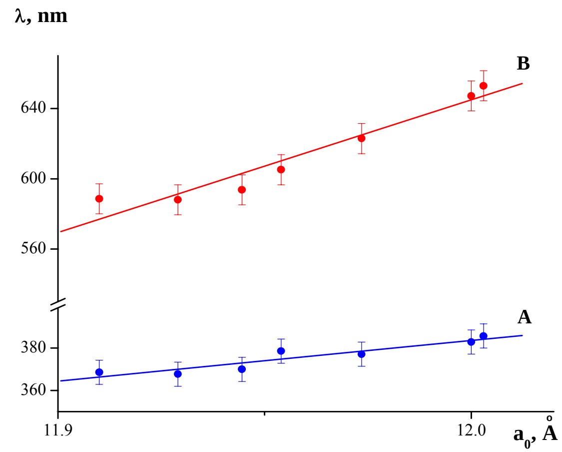


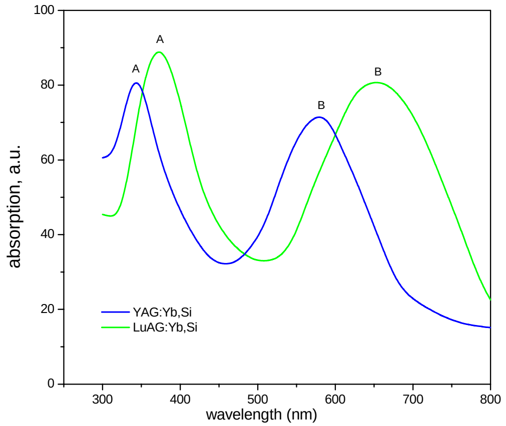


Radiation induced centers in LuAG



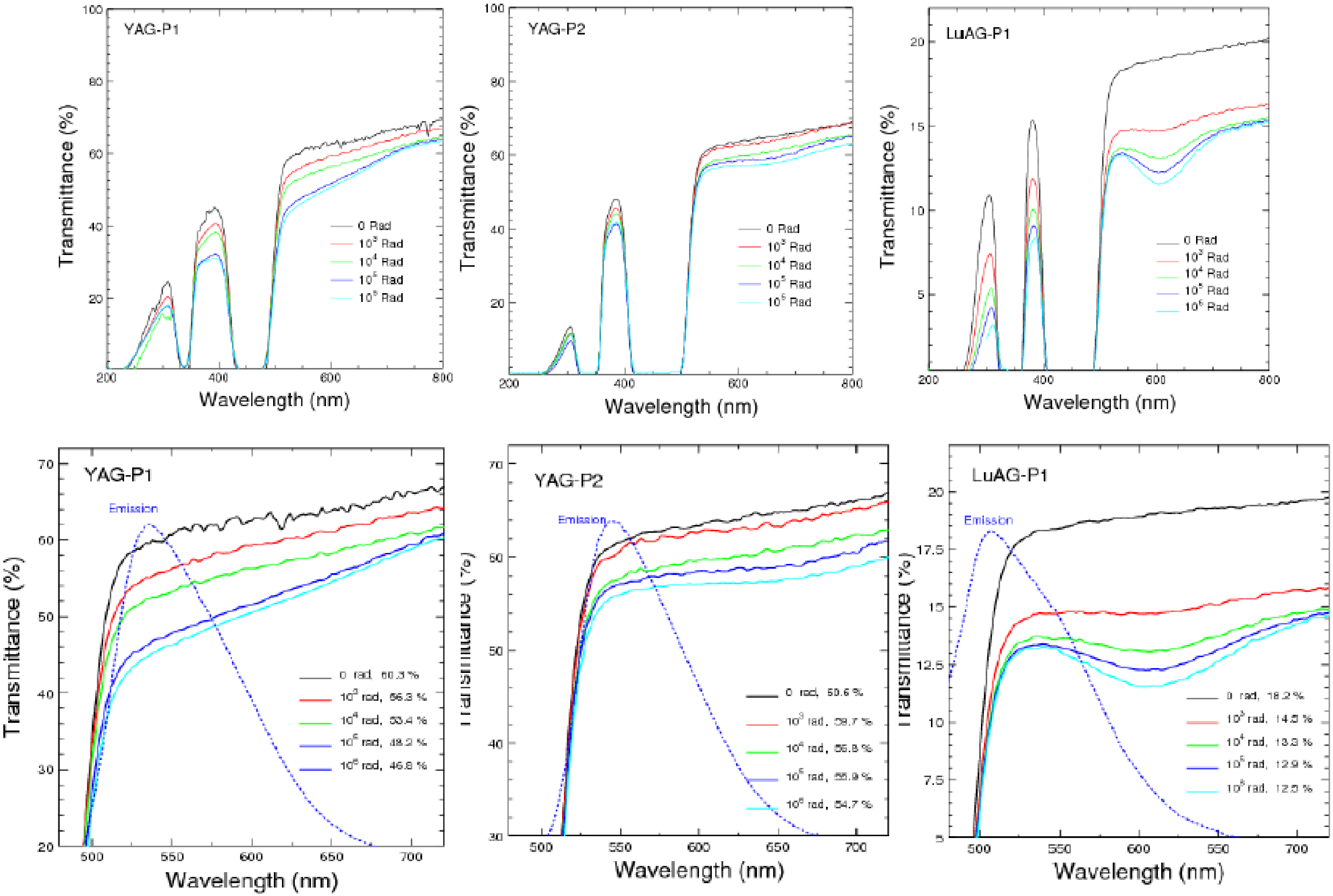
Spectral locations of gamma-ray Induced absorption bands in (Lu,Y)AG:Ce





Spectral locations of Yb^{2+} bands in rare-earth garnets as a function of the lattice parameter
(T.I.Butaeva, A.G.Petrosyan, A.K.Petrosyan, Inorganic Materials 24, 1988, 430)

ceramics



YAG-P1
YAG-P2
LuAG-P1
Transmittance (%)
Wavelength (nm)
0 Rad
10^3 Rad
10^4 Rad
10^5 Rad
10^6 Rad
Emission
0 rad, 60.3 %
10^3 rad, 56.3 %
10^4 rad, 53.4 %
10^5 rad, 48.2 %
10^6 rad, 46.8 %
0 rad, 60.6 %
10^3 rad, 58.7 %
10^4 rad, 56.8 %
10^5 rad, 55.9 %
10^6 rad, 54.7 %
0 rad, 18.2 %
10^3 rad, 14.5 %
10^4 rad, 13.8 %
10^5 rad, 12.9 %
10^6 rad, 12.5 %

Institute for Physical Research

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IPR's scientific activities span:

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- Quantum information
- Photonics and microstructured materials
- Interaction of radiation with matter
- New solid-state laser materials and schemes
- New scintillation materials
- Growth and characterization of laser and scintillation crystals
- Thin film structures for microelectronics and laser technologies
- Solid state physics; organic ferromagnetism
- High-temperature superconductivity
- Synthesis and characterization of nanomaterials
- Scientific instrumentation

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- Laboratory of Quantum Informatics
- Physics Engineering Laboratory
- Laboratory of Solid State Lasers and Spectroscopy
  - Coating Facility
- Laboratory of Crystal Growth of Luminescence Materials
  - X-ray Analysis
- Laboratory of Non-Linear Crystals and Elaborations
- Laboratory of Crystal Optics
  - γ-Radiation Station
- Laboratory of Solid State Physics
- Laboratory of High-Temperature Superconductivity
  - Electron Microscopy and X-ray Microanalysis
- Laboratory of Superconducting Detectors' Physics

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Candidates: 43
PhD students : 9
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In a region rich with historical monuments.

7th century church

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Poster sessions organized by young participants in the Institute's apricot garden.