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CRYSTAL CLEAR

Multipurpose scintillation materials

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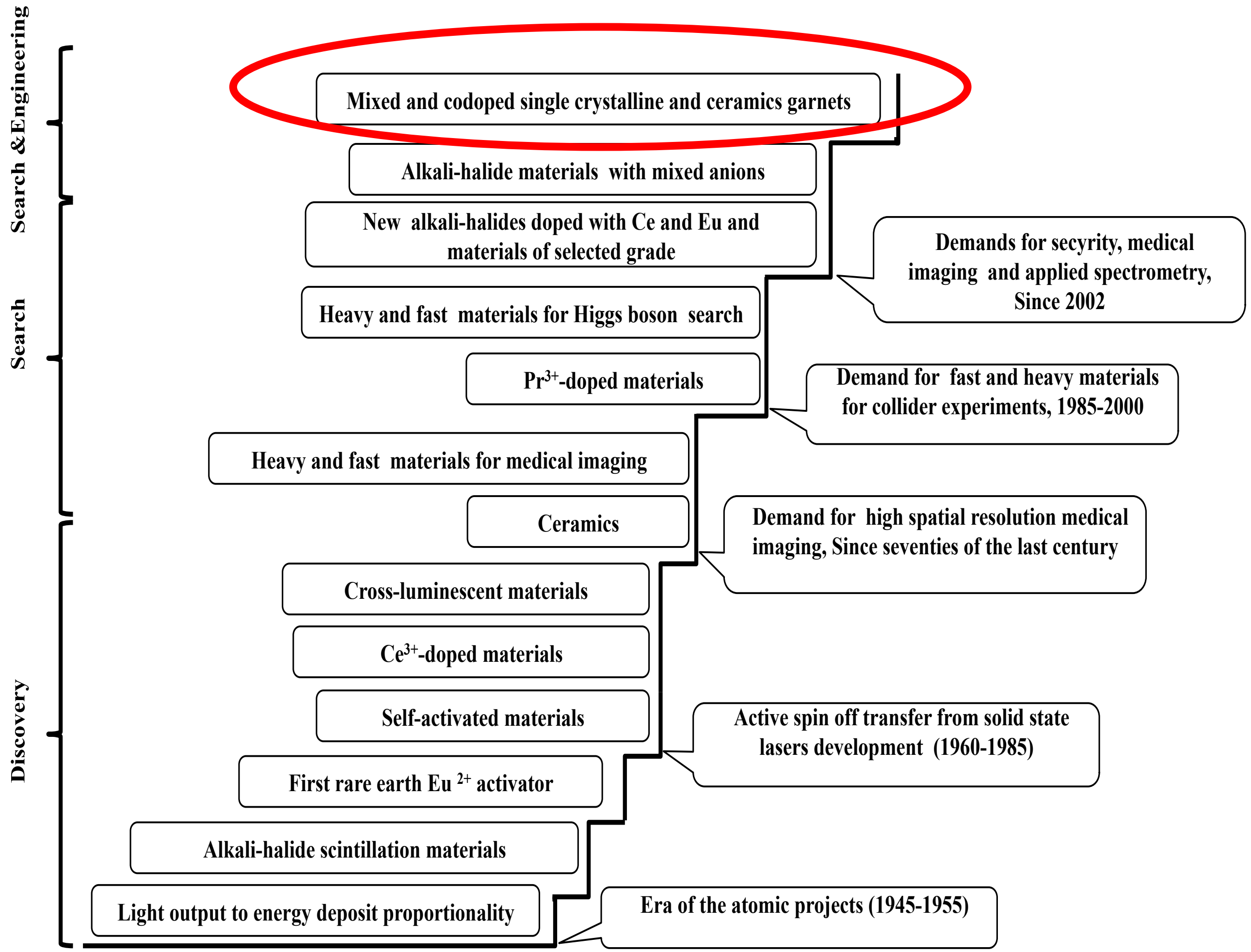
(This work is along the line of the Crystal Clear Collaboration (CERN) and is supported by Russian Federation Government grant number 14.W03.31.0004.

Outline

- What does mean multipurpose scintillator?
- Disordered crystalline materials
- Demand for a fast timing at the conditions of a harsh irradiation environment of experiments at colliders
- Energy release and scintillation photon rate production in different configurations of the detecting modules
- Coincidence time resolution with LYSO and GAGG scintillators
- Demand for neutron detection
- Advertising

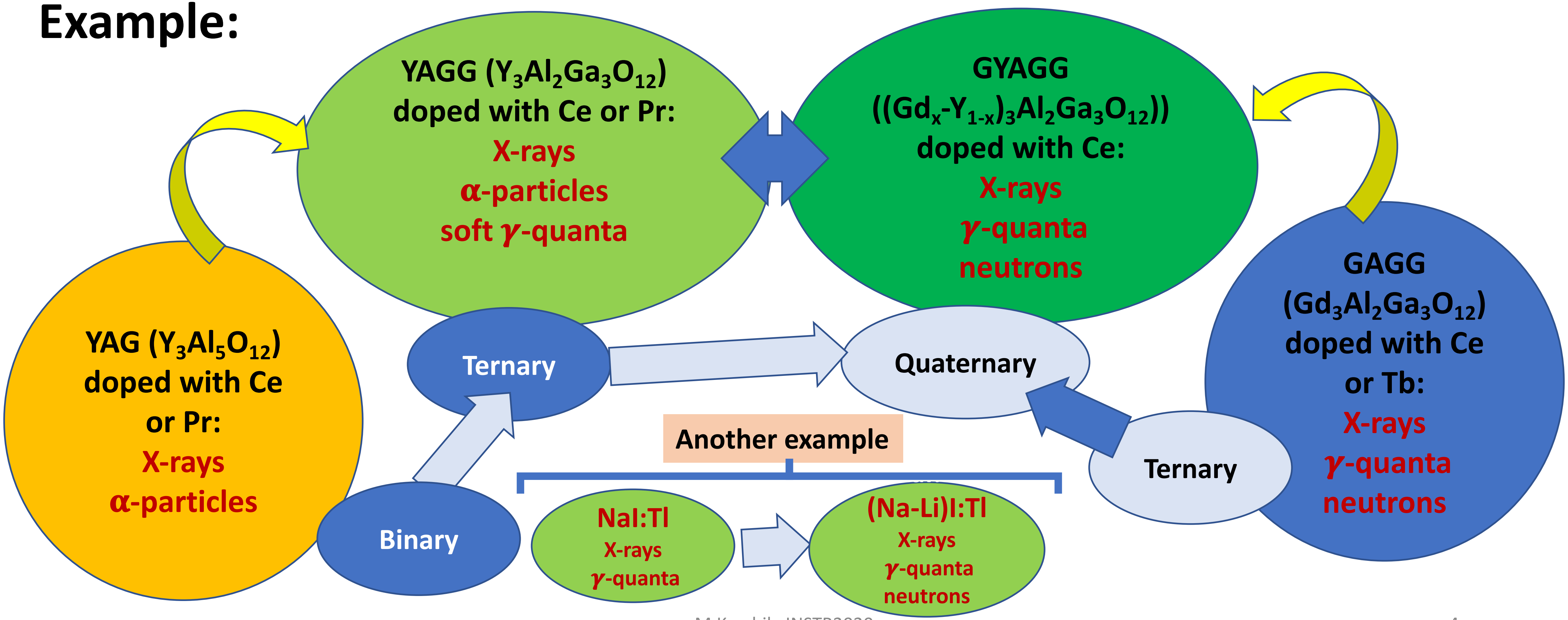
History of scintillators development



Multipurpose scintillation materials

Materials allowing at the reshuffling of their composition a detection of different kinds of the ionizing radiation

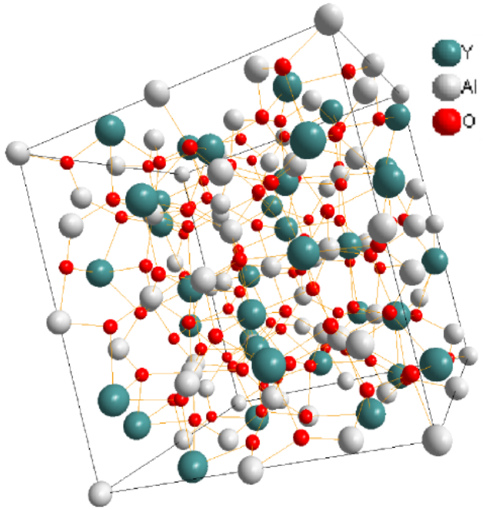
Example:



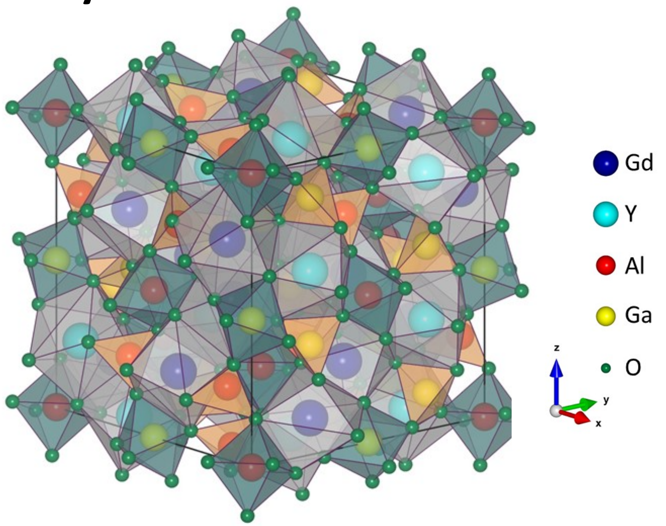
Disordered crystalline materials

A mixture of the different kinds of cations in the crystal increases a diordering in the crystal

Ordered $Y_3Al_5O_{12}$
single crystalline scintillation material



Disordered $(Gd_{0.5}\text{-}Y_{0.5})_3Al_2Ga_3O_{12}$
single crystalline scintillation material

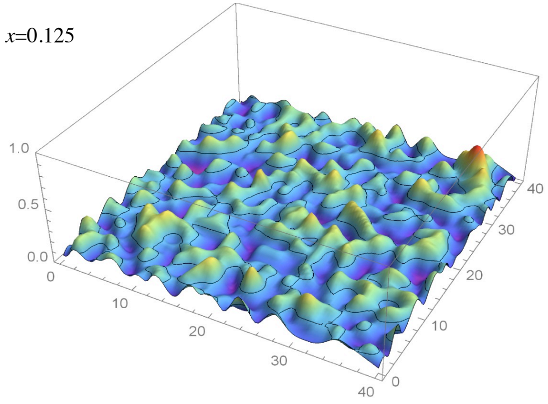


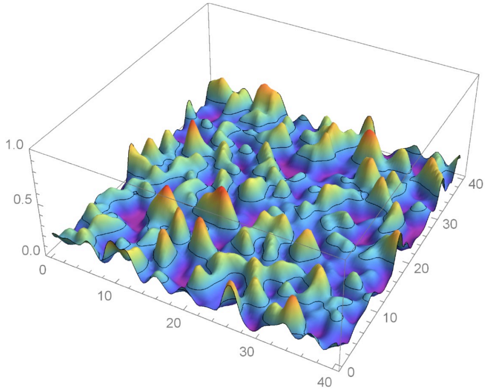
(!) Cationic subblatice is disordered, but Anionic lattice is ordered
Consiquences:

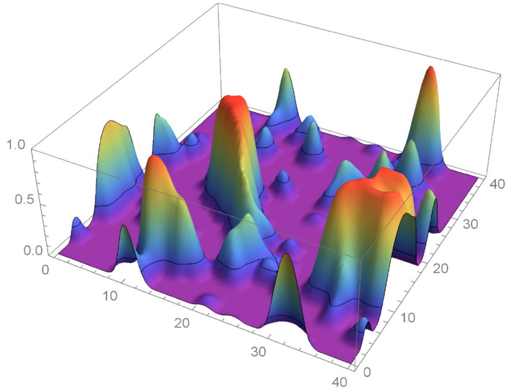
1. **The summetry porperties remain the same;**
2. **The crystalline field porperies and effects porviding the energy levels structure remain the same;**
3. **The dynamics of nonequlibrium carriers and energy transfer porcesses become differrnt.**

Disordering to reduce a scattering length of excitons and free carriers in scintillator

Variation of different cations in the matrix allows an engineering of the conduction bottom landscape to control parameters of the track of the nonequilibrium carriers



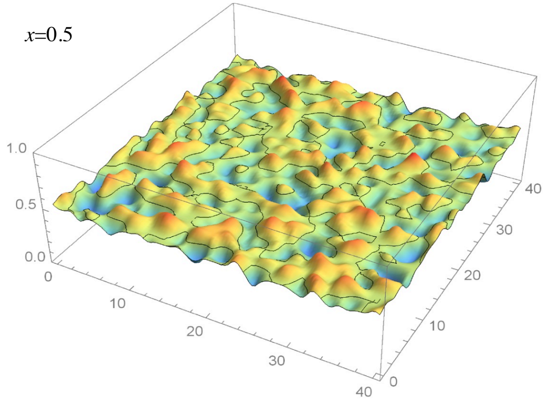


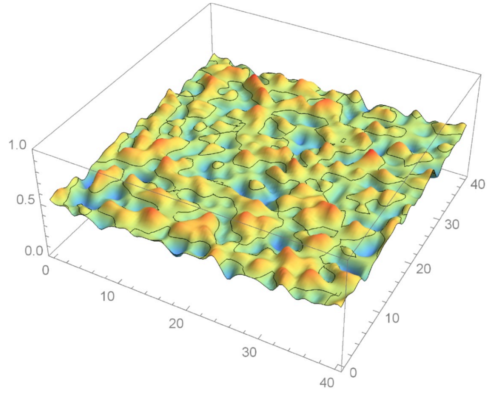


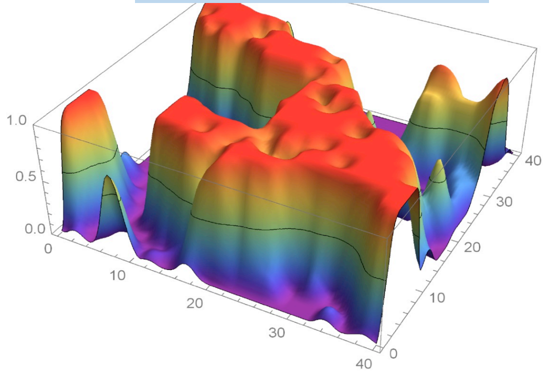
$U_{AA}=U_{BB}>U_{AB}$

$U_{AA}=U_{BB}=U_{AB}$

$U_{AA}=U_{BB}<U_{AB}$







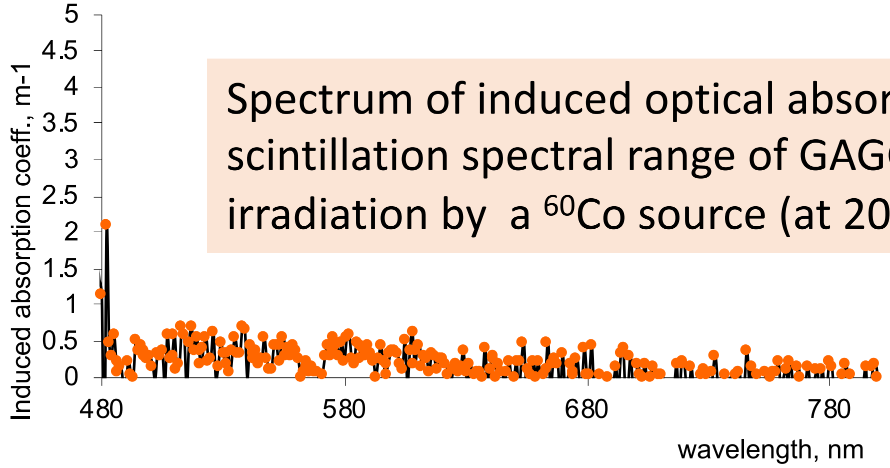
Simulation of the conduction bottom landscape in binary systems

Disordering in the matrix promotes an improvement of the scintillation parameters

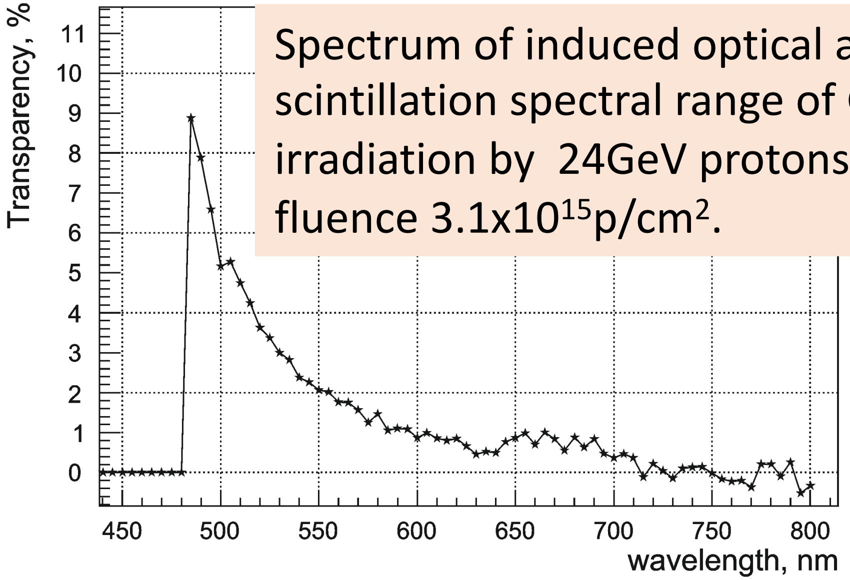
	YAG **$(Y_3Al_5O_{12})$:Ce**	**YAGG** **$Y_3Al_2Ga_3O_{12}$:Ce**	**GYAGG** **$(Gd_x\text{-}Y_{1-x})_3Al_2Ga_3O_{12}$): Ce,Mg**	**GAGG** **$Gd_3Al_2Ga_3O_{12}$: Ce,Mg**
LY, ph/MeV	30000	20000	50000	41000
Decay/fraction ns(%)	80	37	36 (80) 97 (20)	28 (30) 68 (52) 168 (18)
Radiation tolerance **γ-quanta** **hadrons**	🤓 🤓	🤓 Not studied	🤓 Not studied	🤓 🤓
Scintillation maximum, nm	550	530	510	520

Disordering in the matice keeps the tolerance to irradiation. GAGG meets requirents for irradiation loading of HL LHC and coming FCC experiments

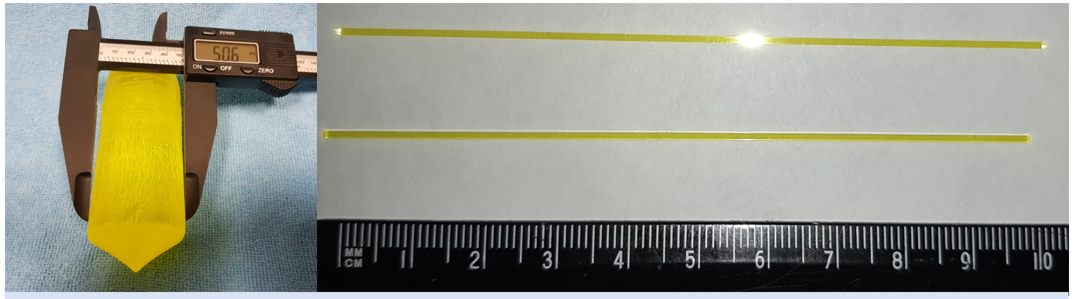
Crystal	GAGG:Ce,Mg Ti (multidoped GAGG)
Scintillation decay constants, ns, and relative weight, (%)	28 (30) 68 (52) 168 (18)
Light yeird, Ph/MeV	41000
Coincidence time resolution for 511 keV, ps	160
dE/dx, mip per mm of the media, MeV	0.78



Spectrum of induced optical absorption in scintillation spectral range of GAGG after irradiation by a ^{60}Co source (at 2000Gy).



Spectrum of induced optical absorption in scintillation spectral range of GAGG after irradiation by 24GeV protons with fluence $3.1 \times 10^{15} p/cm^2$.



$Gd_3Al_2Ga_3O_{12}$:Ce,Mg,Ti-multidoped GAGG scintillation crystal is under mass production by FOMOS CRYSTALS (Moscow Russia)

M.Korzhik, INSTR2020

Demand for the scintillation materials providing good timing porperties for experiements at colliders

Requirments to the scintillation materils:

- Tolerance to irradiation combineed γ-quanta and hadrons;
- Fast leading edge of the scintillation pulse;
- Short decay time comparable with duration between bunches;
- Hight light yield porviding large amplitude of the scintillation pulse;
- Good scintillation attenuation length providing an ability to use relatively long crystalline elements like in ”spocal” design;

A requirement forever:

- Existing capacity for production to minimise expenses at R&D stage

KINETICS PARAMETRS OF LUMINESCENCE RESPONSE ARE THE KEY PARAMETERS IN REALLY FAST SCINTILATION DETECTORS

The mean delay of the timestamp of an interaction event $\langle t_{st} \rangle$
for registration with threshold N_{th}
for scintillation materials of different types

Decay time

Instantaneous response
(cross-luminescent scintillators, e.g., BaF_2)

$$\langle t_{\mathrm{st}} \rangle \approx \tau_{\mathrm{d}} \frac{N_{\mathrm{th}}}{N_{\mathrm{em}}}$$

Rise time

Fast response
(selfactivated scintillators, e.g., PWO, BGO)

$$\langle t_{\mathrm{st}} \rangle \approx \sqrt{\tau_{\mathrm{d}} \tau_{\mathrm{r}} \frac{N_{\mathrm{th}}}{N_{\mathrm{em}}}}$$

IMPORTANT FOR REALLY FAST ACTIVATED SCINTILLATORS

Exitation transfer time

Delayed response
(activated scintillators, e.g., LSO, LYSO, GAGG)

$$\boxed{\langle t_{\mathrm{st}} \rangle \approx \left(\frac{3\tau_{\mathrm{d}} \sqrt{\tau_{\mathrm{ex}}}}{4q} \frac{N_{\mathrm{th}}}{N_{\mathrm{em}}} \right)^{2/3}}$$

G.Tamulaitis, A.Vasil'ev, M Korzhik, A Mazzi, A Gola , S.Nargelas, A Vaitkevičius , A.Fedorov, and D.Kozlov, Improvement of the Time Resolution of Radiation Detectors Based on Gd3Al2Ga3O12 Scintillators with SiPM Readout, IEEE Trans. Nucl. Sci. **66**, 1879 (2019)

Contribution of the scintillator in threshold. Radioluminescence due to radioisotopes

Short and long living radio-isotopes created in the material under irradiation conditions at HL LHC

Contribution of harmful luminescence in optical noise, noise pedestal, and noise energy equivalent within 25 ns gate in GAGG and LSO crystals with dimensions of $2x2x0.2\ cm^3$ [*]

Scintillator	GAGG	LSO
Additional noise intensity, ph	1600	1350
Additional pedestal, keV	156	108
Additional noise energy equivalent, keV	9.2	6.9

*Auffray A, Dosovitskiy G, Fedorov A et.al. (2019) Irradiation effects on Gd3Al2Ga3O12 scintillators prospective for application in harsh irradiation environments Radiation Physics and Chemistry, V164:108365

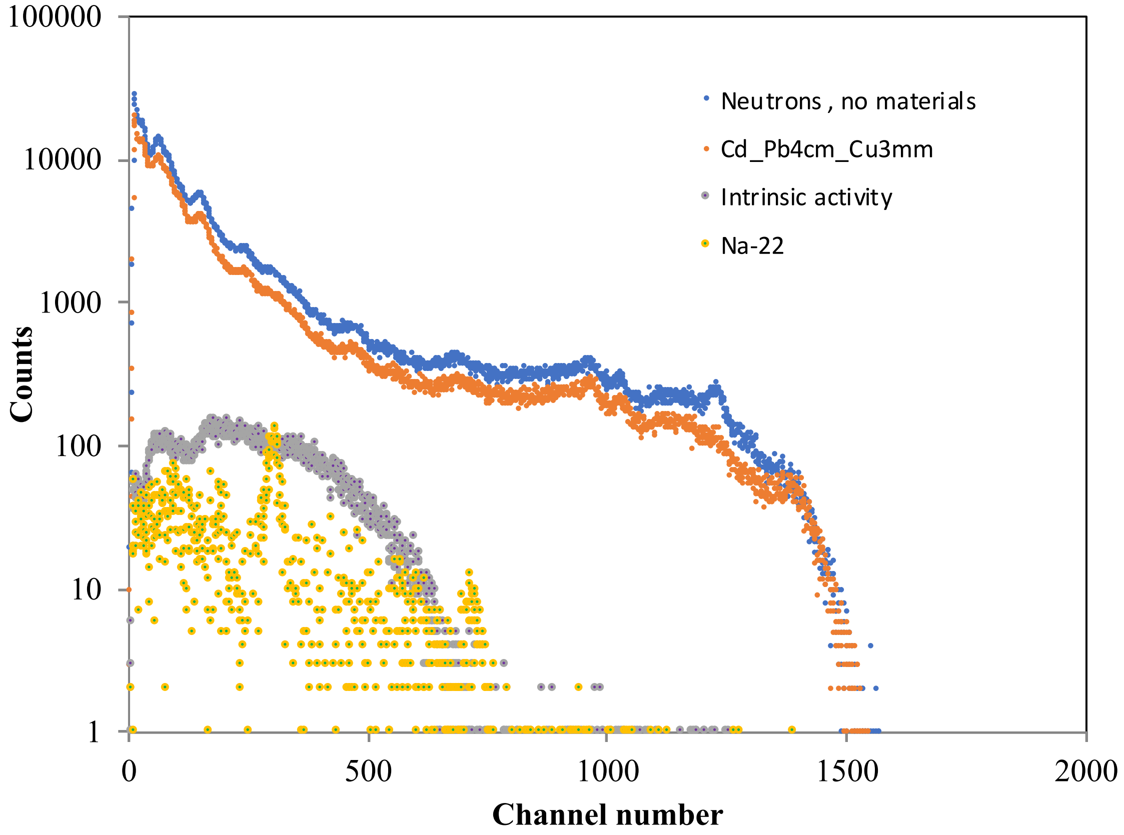
Contribution of the scintillator in threshold. Neutrons

Neutron radiative capture cross section σ_γ and radiative capture resonance integral I_γ

Element	σ_γ, b	I_γ, b
Gd	49000	390
Ga	2.9	18.7
Ce	0.63	3.0
Lu	77	900
La	9.0	12.2
Br	6.8	90
O	2.7E-4	3.1E-4
Al	0.23	0.17
Si	0.16	0.08

Soft γ-quanta spectra created under neutrons of Am-Be source in LYSO and detected with the same scintillator

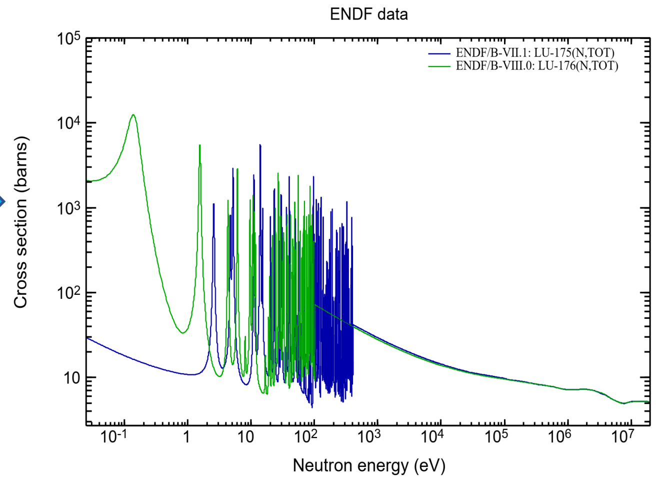
The γ- quanta spectra measured with LYSO:Ce 10x10×2 mm^3 sample.



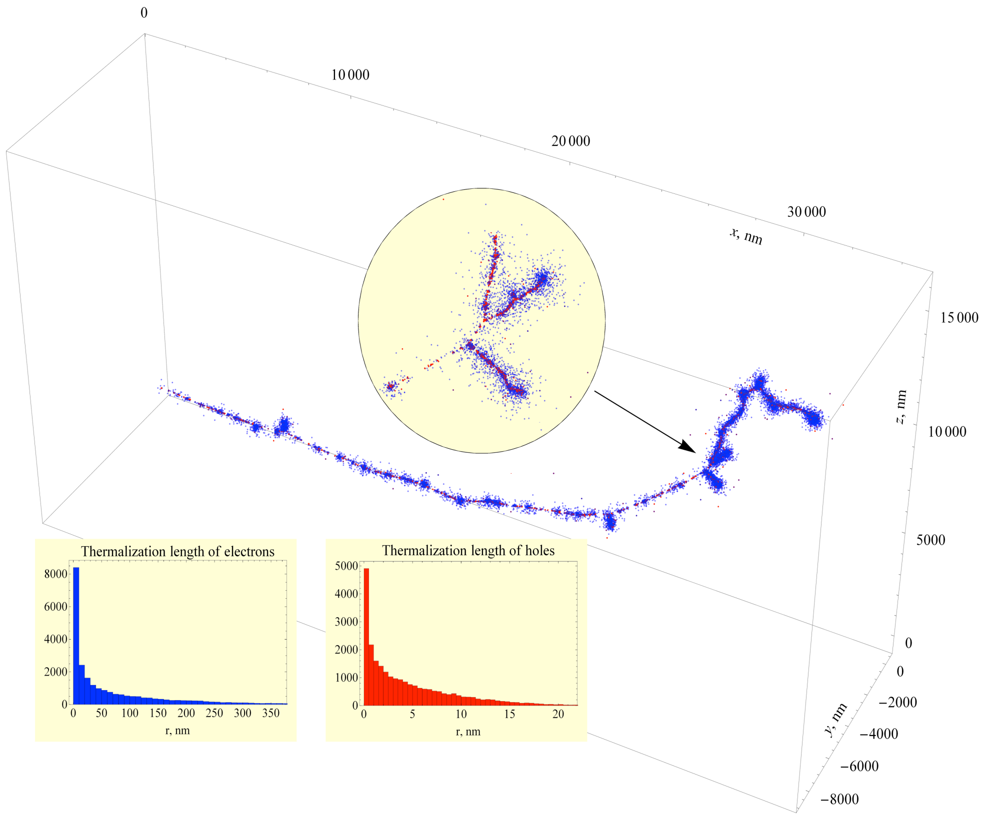
Neutron radiative capture cross section σ_γ and radiative capture resonance integral I_γ

Element	σ_γ, b	I_γ, b
Lu	77	900
O	2.7E-4	3.1E-4
Si	0.16	0.08

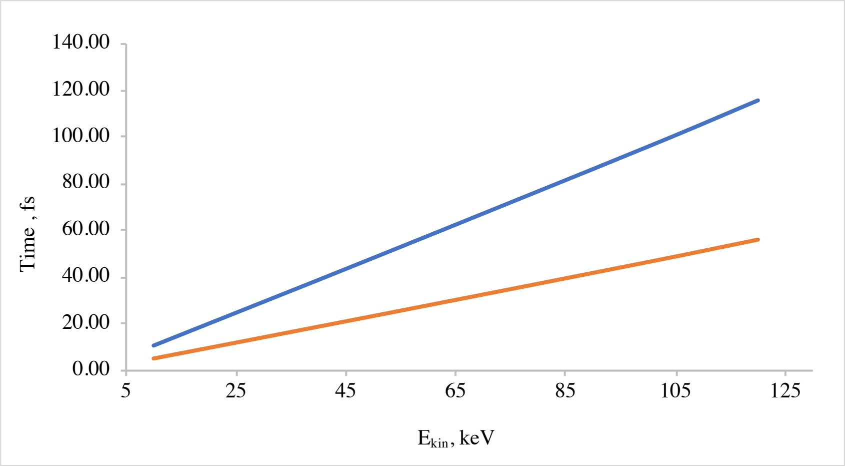
Cross sections on isotopes of Lu in epithermal and fast region.



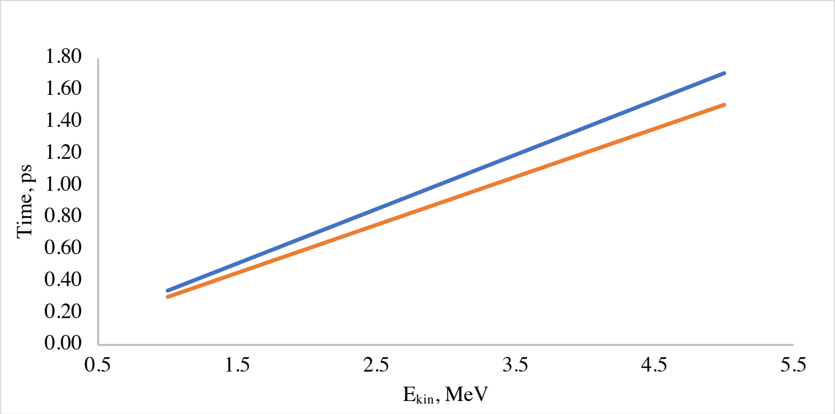
Energy release in the scintillation material



Spatial distribution of electrons and holes after thermalization due to photo-absorption of a 200 keV γ-quantum in NaI(Tl) scintillator

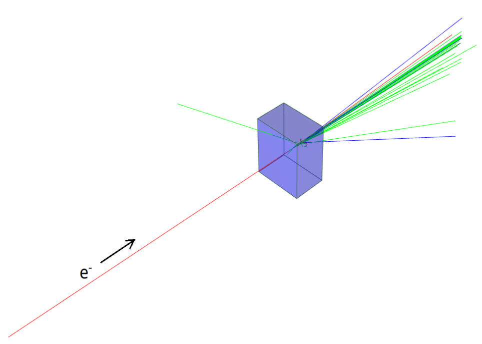


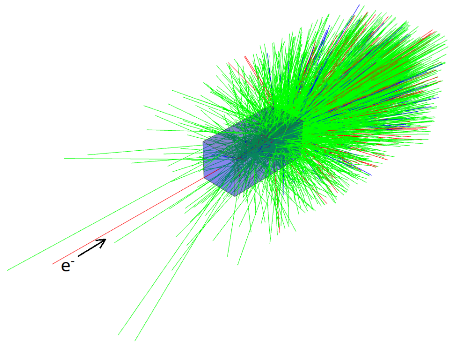
Dependence of the average energy release time on kinetic energy of a non-relativistic electron in light and heavy scintillators: $Y_3Al_5O_{12}$: Ce (blue) and $PbWO_4$ (red)

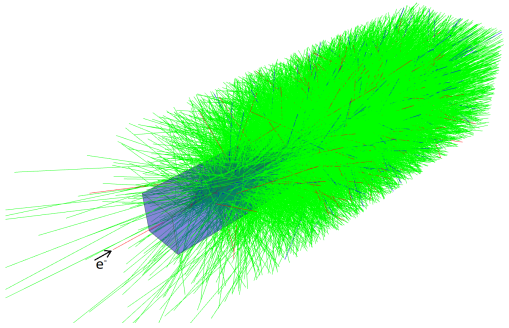


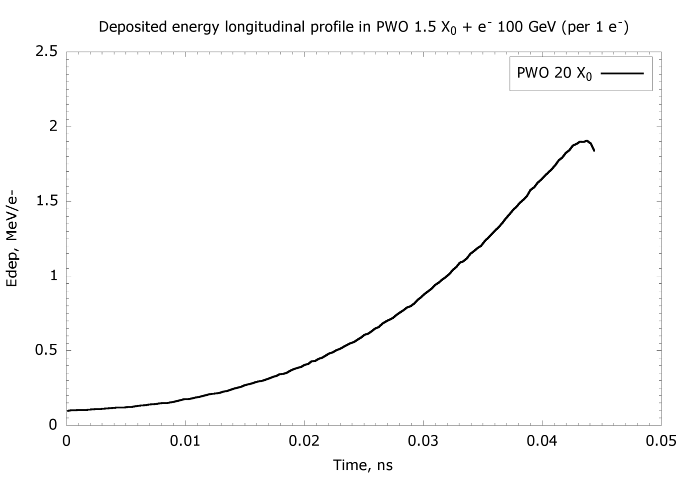
Dependence of the average energy release time on the kinetic energy of alpha -particle in two scintillators: $Y_3Al_5O_{12}$: Ce (blue) and $PbWO_4$ (red)

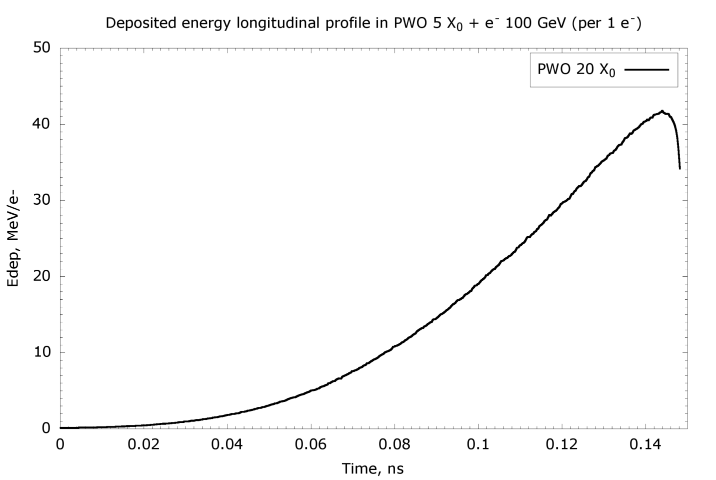
GEANT4 image of interaction products (upper panels) and time profile (bottom panels) for energy deposit of a 100 GeV-electron in lead tungstate scintillation crystal with a cross section of 2x2 cm^2 and a length of 1.5, 5 and 20 X_0

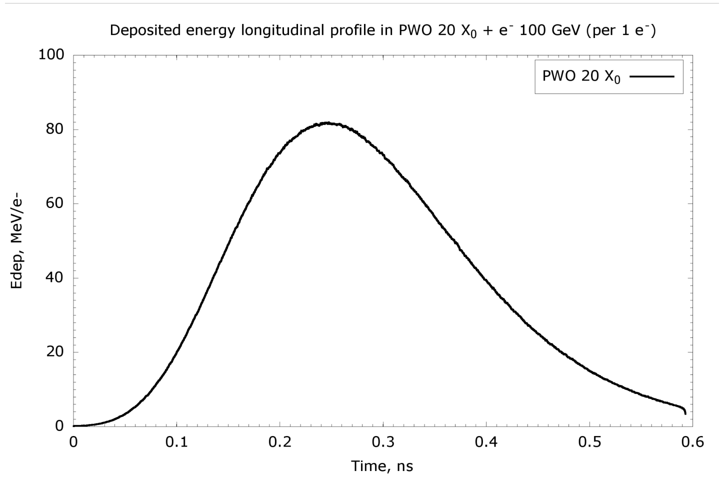






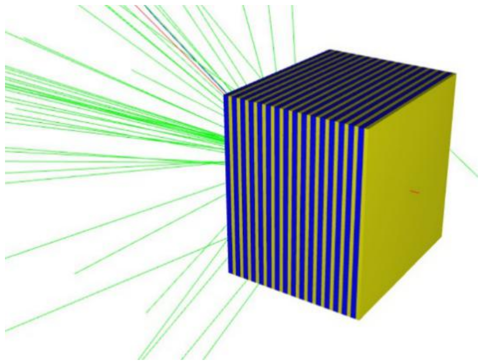


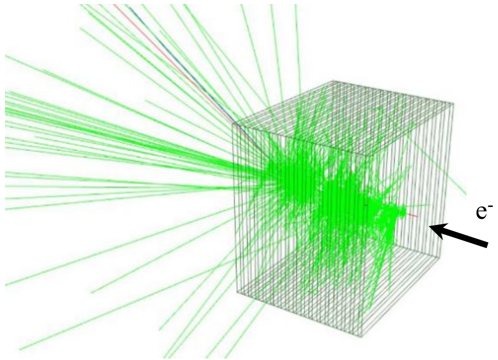


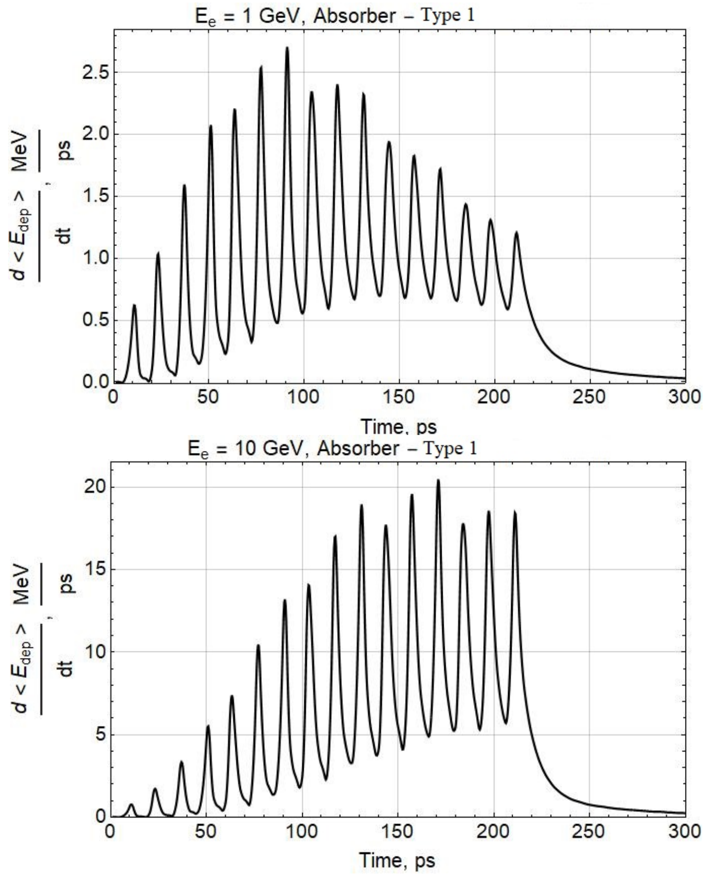


Energy release time is a limiting factor in a long homogeneous scintillator detecting cell for a fast timing!

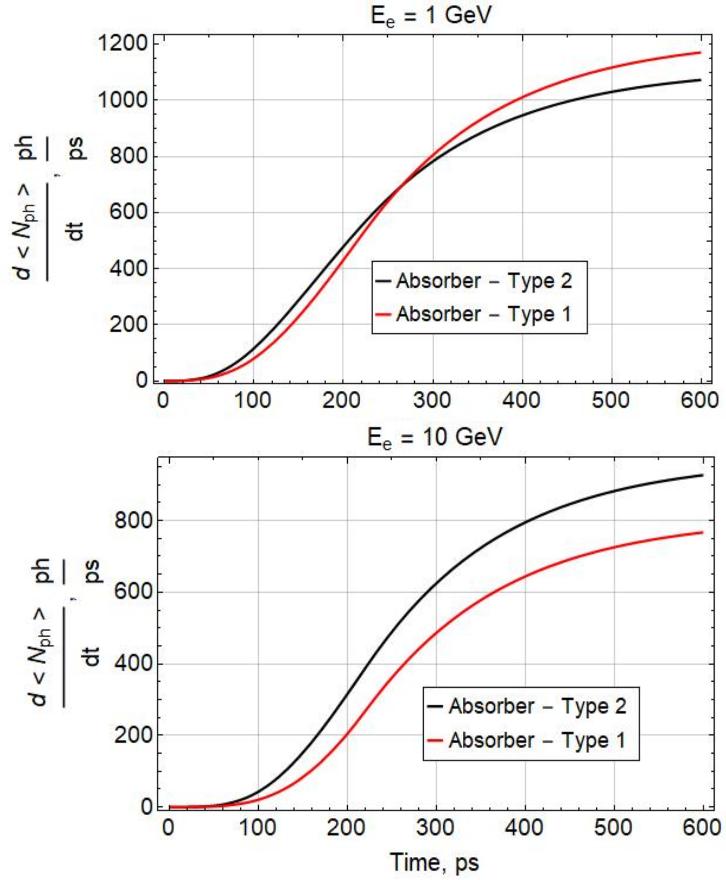
GEANT4 simulation of the energy release and the scintillation photons emission rate in the "shaslyk" type module consisted of 2mm W/Cu(75/25%) plates and 2 mm GAGG plates





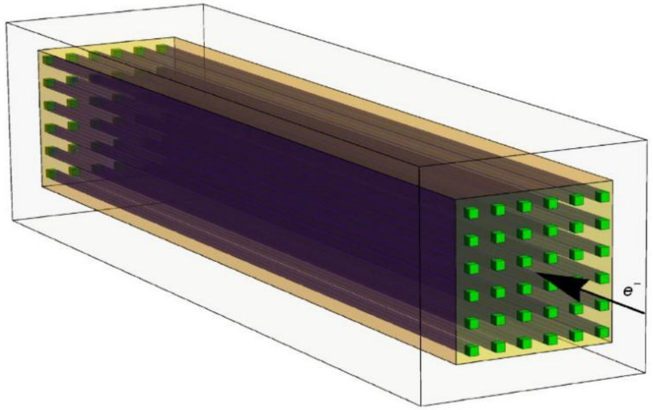


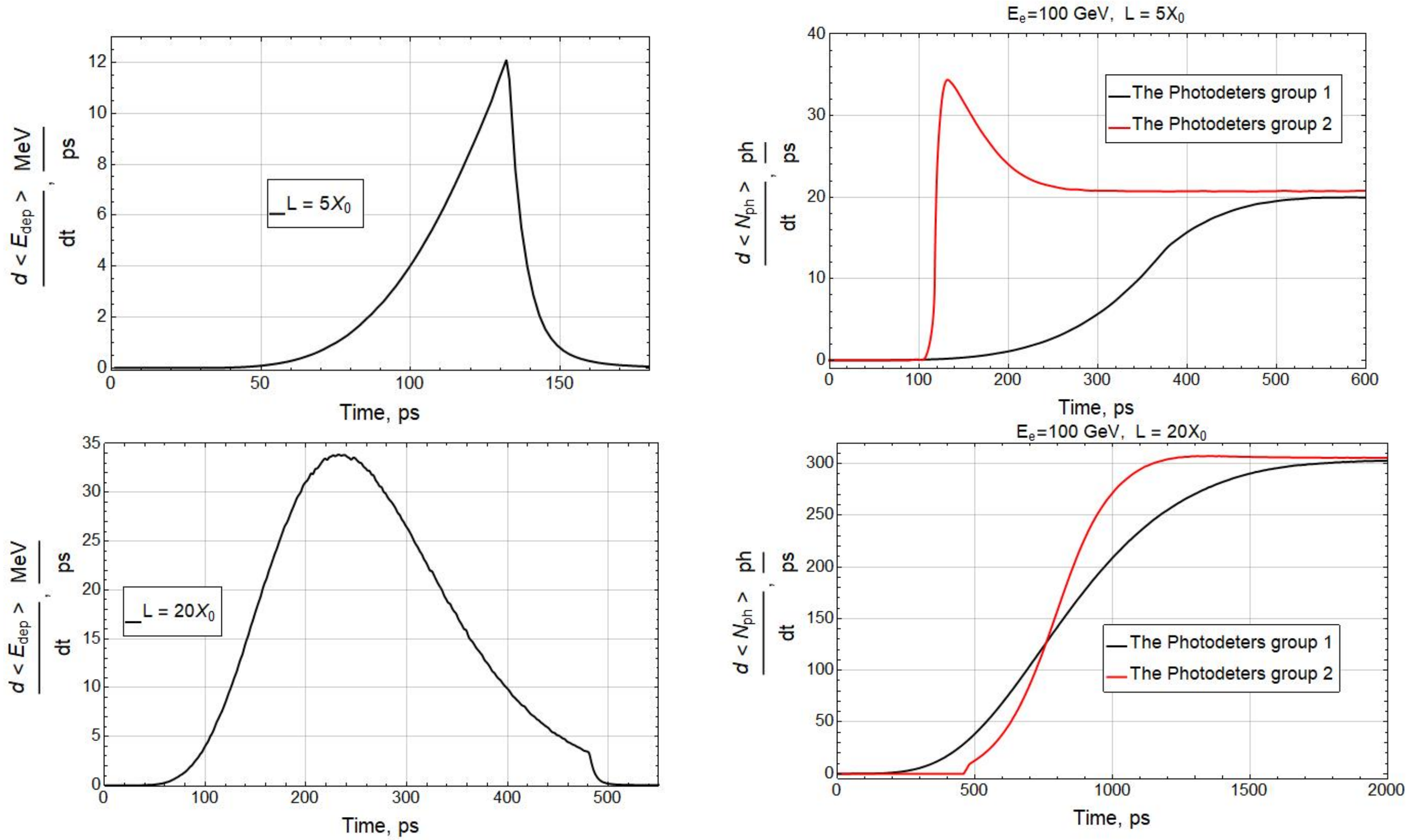
The average energy release rate in all GAGG scintillation plates at the electron energy of 1 GeV (top) and 10 GeV (bottom)



The average photon emission rate in all scintillation plates for two types of absorbers at the electron energy of 1 GeV (top) and 10 GeV (bottom). Type 1- W/Cu(75/25%), Type 2- pure W.

GEANT4 simulation of the energy deposit and scintillation photons arrival rate in the “spacal” type module consisted of $2x2xNX_0$ mm GAGG fibers and W/Cu(75/25%) absorber





The average energy release rate in all fibers for a spacal cell at its lengths of 5X0 (top) and 20X0 (bottom)

The average photon arrival rate in all fibers for a spacal cell at its lengths of 5X0 (top) and 20X0 (bottom)

Coinsidence time resolution with small size scintillation elements and annihilation γ-quanta

Measuremnts can be easy used to estimate response to MIPs

Material	Plastic	$Y_3Al_5O_{12}$: Ce,Mg (YAG)	Lu_2SiO_5: Ce (LSO:Ce)	$(Lu_x\text{-}Y_{1-x})_2SiO_5$:Ce (LYSO:Ce)	$Gd_3Al_2Ga_3O_{12}$ (GAGG) multidoped
ρ, g/cm³	1	4.55	7.4	7.2	6.63
LY, ph/MeV	10000	30000	27000	29000	46000
dE/dx @ e^-, π, MeV/mm	0.158	0.59	0.87	0.85	0.81
Yield, ph per 1 mm per mip	1580	17700	23500	24600	37200

Disordered scintillation materials are better with an additional cooping

An effect of the Ca cooping on the CTR of LYSO:Ce

FWHM (in ps) in coincidence time resolution (CTR) measurements at different temperatures measured with 3x3x5 mm samples and NUV HD SiPM (FBK, Trento)

Sample	LSO	LYSO:Ce	LYSO:Ce,Ca	LYSO:Ce, Ca
20°C	115+/-1	105+/-1	93+/-1	93+/-1
0°C	117+/-2	106+/-1	95+/-2	93+/-1
-20°C	121+/-1	109+/-1	104+/-2	101+/-2

Decay time of scintillation kinetics at room temperature

Sample	LSO	LYSO:Ce	LYSO:Ce,Ca	LYSO:Ce, Ca
τ_d, ns	40	39	39	37

Ca cooping concentration at the level of 5-10 ppm:

1. makes CTR low dependent on Ce concentration;

2. Makes CTR less dependent on technological variation of kinetics decay time;

Disordered scintillation materials are better with an additional cooping to Ce

An effect of the Mg cooping on the CTR of GAGG:Ce, Ti

FWHM (in ps) in coincidence time resolution (CTR) measurements at different temperatures measured with 3x3x5 mm samples and RGB HD SiPM (FBK, Trento)

Sample	CTR FWHM, ps, at different temperatures		
	+20^{o}C	0^{o}C	-20^{o}C
GAGG:Ce, Ti.Mg	165±3	160±2	164±2
GAGG:Ce	210±3		

Demand for neutron detection tewchniques

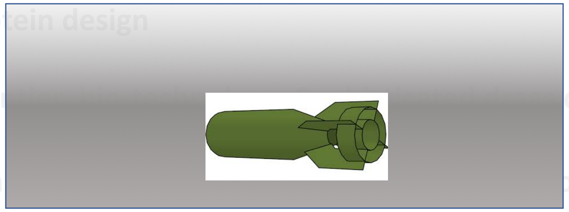
1. Neutron is a massive particle; therefore its De Broglie wavelength is sufficiently shorter even at a small kinetic energy

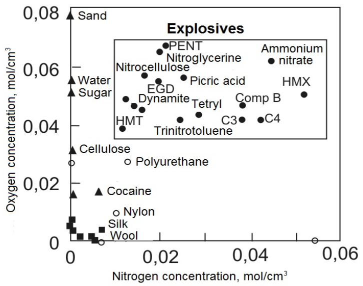
2. Mechanism of interaction with nuclei is diverse and, again, depends on the De Broglie wavelength of the particle

3. Cross-section of the neutron interaction with nuclei of light atoms of the matter is quite substantial in opposite to gamma-quanta interacting with their electron shells

4. In a dense matter, neutrons have sufficiently longer trajectories in a comparison with charged particles and X-rays and gamma-quanta

Remote detection of substances consisted of light atoms





Remote inspection of explosives and fissile materials

Inspection of the ground for the "unexpected explosive gifts" from the WW-II

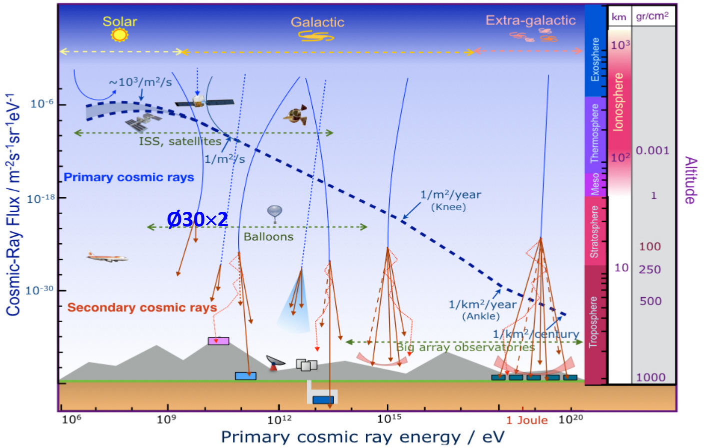
Hydrocarbon search and wells exploration

Neutrons from the space around the Earth

Solar neutrons penetration

(white jets) (blue sprites) (elves)

Sensors at satellites or meteorological missiles are the most reliable tools to probe neutrons from the space



(See more details A.F. Iyudin, S.I. Svertilov in Eds. M.Korzhik, A.Gektin , Engineering of Scintillation Materials and Radiation Technologies , Springer, 2019)

Neutron discrimination for better antineutrino detection

Remote inspection of the production cycle at the nuclear energy plants

$$\bar{\nu}_e + p \rightarrow e^+ + n$$

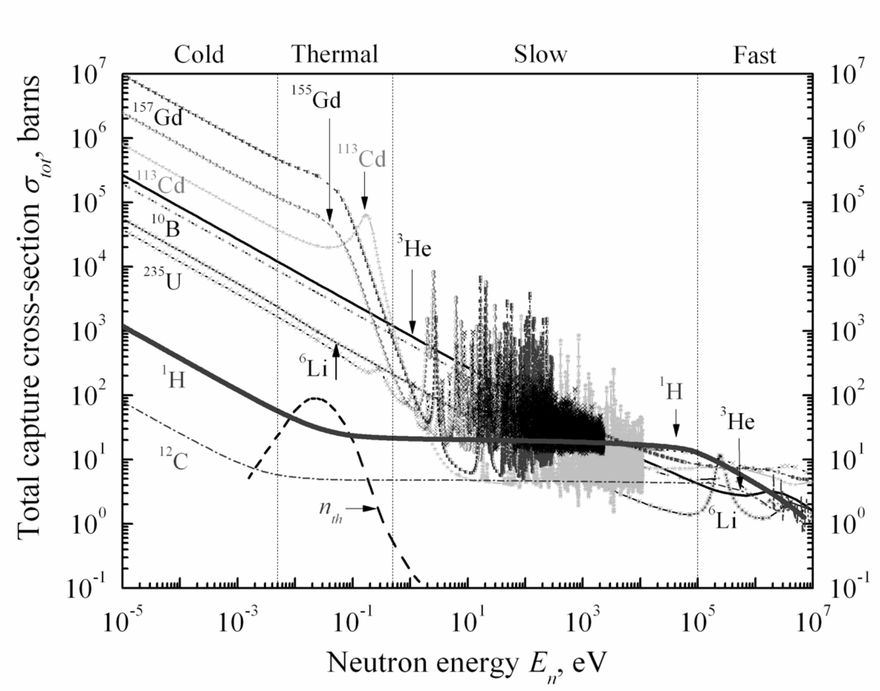
Neutrino detection mobile module

A..Hightighat et.al., Observation of reactor antineutrinos with a Rapidly-Deployable surface level detector, arXiv:1812.02163v1 [physics.ins-det] 5 Dec 2018

Limiting factors to construct neutron sensitive inorganic scintillation materials

Interaction with neutrons

Neutrons with low kinetic energies can be detected with light nuclei containing media with electric readout (^{3}He gas counters) or optical readout (^{6}Li, ^{10}B) through scintillation



Concerns:

- **Concentration is limited to $5*10^{20}$ atoms cm^{-3} in ^{3}He counters;**
- **^{3}He lack at the marked;**
- **No bright B based**
- **scintillators are discovered yet;**
- **^{6}Li enrichment is expensive**

Scintillation materials to detect fast neutrons

Material	Products of interaction	Advantages Disadvantages
Organic plastic scintillator	Recoil protons	• Cheap technology • Escape of the recoil protons • Low Stokes shift for emitting center • Radiation damage of matrix& luminescent center • Low thermo-stability
Organic crystals	Recoil protons	• Better density compared to plastics;
Organic glass	Recoil protons	• New prospective direction of technology;
Inorganic glass	α,t,γ	• Cheap technology (except ^{6}Li raw material); • Low light yield at **Ce^{3+}** doping • High light yield at **Tb^{3+}** doping, but slow
ZnS(Ag)LiF composites	α,t	• Large area detectors • Low count rate
Inorganic crystalline materials	α,t,γ	• High light yield ; • **High Z_{eff} → high photo-absorption** Need for (α,t)/γ discrimination

Products of interaction with neutrons

Fissile materials are out of interest

Gamma-quanta under 14MeV neutron source

Element	Reaction	Cross section (mb)	E_γ, MeV
C	(n,n’gamma)	200	4.44
O	(n,n'gamma)	750	6.13
N	(n, gamma)	75	10.83
Cl	(n, gamma)	4300	6.11
S	(n, gamma)	520	5.42
H	(n, gamma)	200	2,223

Usually, inorganic scintillators are hydrogen poor

Thresholdless reactions

$$ {}^{6}_{3}Li + n_{therm} \to {}^{3}_{1}H(E_t = 2{,}73 MэB) + {}^{4}_{2}He(E_\alpha = 2{,}05 MэB) $$

$$ {}^{10}_{5}B + n_{therm} \to \begin{cases} {}^{7}_{3}Li + {}^{4}_{2}He(E_\alpha = 1{,}78 MэB), & 6\% \\ {}^{7}_{3}Li + {}^{4}_{2}He(E_\alpha = 1{,}47 MэB) + \gamma(E_\gamma = 0{,}48 MэB), & 94\% \end{cases} $$

Reactions with threshold

$$ {}^{12}_{6}C + n \to {}^{9}_{4}Be + {}^{4}_{2}He - 5{,}702 MэB $$

$$ {}^{12}_{6}C + n \to n + 3\,{}^{4}_{2}He - 7{,}275 MэB $$

$$ {}^{12}_{6}C + n \to {}^{12}_{5}B + p - 12{,}588 MэB $$

$$ {}^{12}_{6}C + n \to {}^{11}_{5}B + {}^{2}_{1}H - 13{,}733 MэB $$

$$ {}^{12}_{6}C + n \to {}^{11}_{5}B + n + p - 15{,}957 MэB $$

Diamond or SiC are very prospective to detect fast neutrons
Concerns: Very small plates available, expensive technology

Potential of inorganic scintillators. Li containing materials

	CLYB	CLYC	CLLC	CLLB	CLLBC
Light yield, gamma, ph/MeV neutron, n/MeV	24 000 90 000	20 000 70 000	35 000 110 000	45 000 150 000	45 000 150 000
Energy Resolution, % @662 keV	4.1	4.0	3.4	2.9	3
Emission, nm	410	370	380	410	410

$$\boxed{{}^{6}_{3}Li + n \rightarrow {}^{4}_{2}He + {}^{3}_{1}H + 4{,}784 MэB}$$

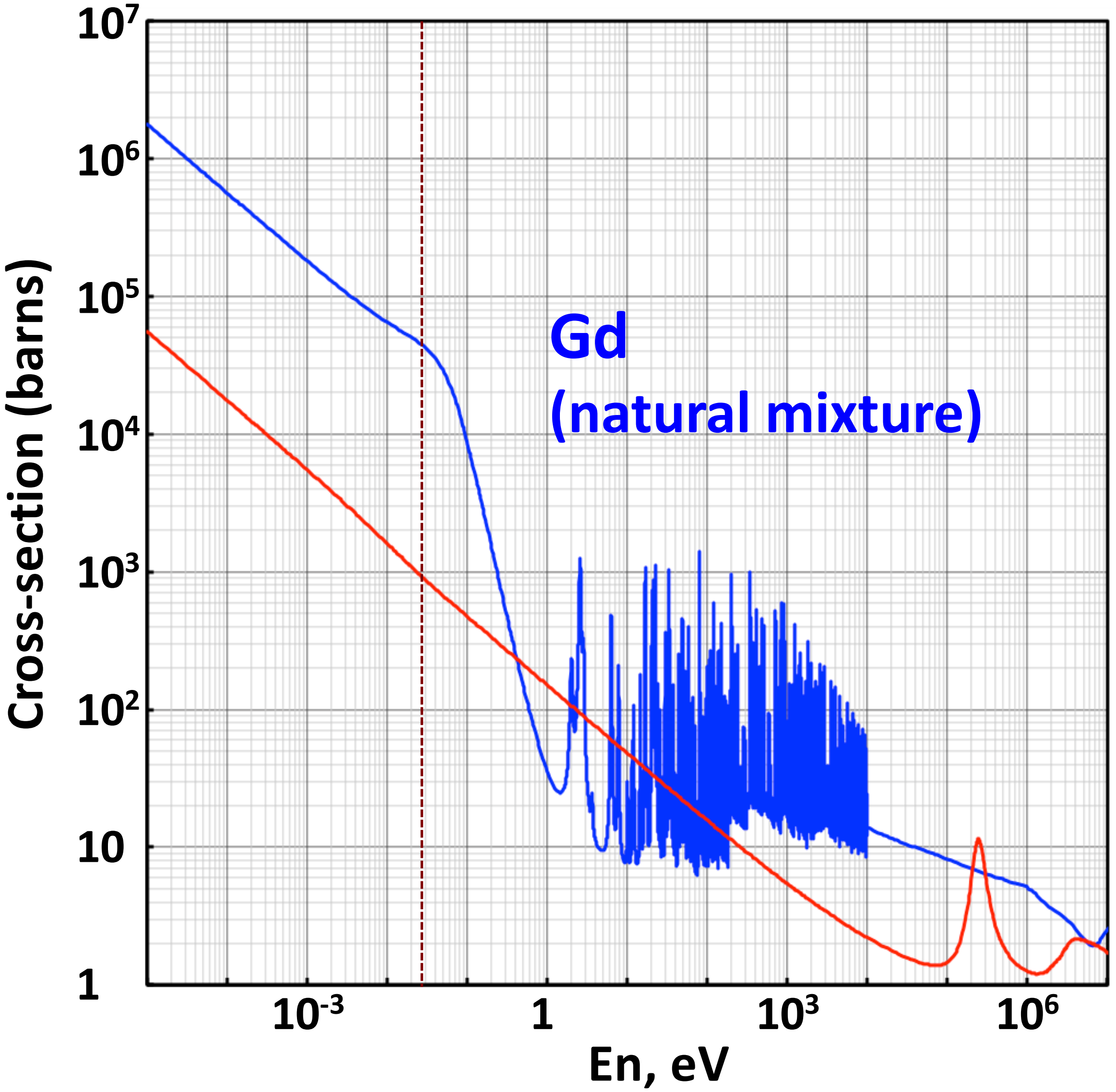
$${}^{6}_{3}Li + n \rightarrow {}^{7}_{3}Li + \gamma + 7{,}25 MэB$$

$${}^{6}_{3}Li + n \rightarrow p + {}^{6}_{2}He - 2{,}727 MэB$$

$${}^{6}_{3}Li + n \rightarrow p + 2n + {}^{4}_{2}He - 3{,}698 MэB$$

(See more details and references in P.Lecoq, A.Gektin, M.Korzhik, Inorganic scintillators , Springer, 2016)

Is Gd superior to lithium?



$$\boxed{{}^{155}_{64}Gd + n \rightarrow {}^{156}_{64}Gd + \gamma + 8{,}536 MэB}$$

$${}^{155}_{64}Gd + n \rightarrow {}^{151}_{62}Sm + n + {}^{4}_{2}He + 0{,}081 MэB$$

$${}^{155}_{64}Gd + n \rightarrow {}^{155}_{63}Eu + p$$

$${}^{155}_{64}Gd + n \rightarrow {}^{152}_{62}Sm + {}^{4}_{2}He$$

$${}^{155}_{64}Gd + n \rightarrow {}^{150}_{62}Sm + 2n + {}^{4}_{2}He - 5{,}515 MэB$$

$${}^{155}_{64}Gd + n \rightarrow {}^{154}_{64}Gd + 2n - 6{,}435 MэB$$

$${}^{155}_{64}Gd + n \rightarrow {}^{154}_{63}Eu + n + p - 7{,}621 MэB$$

$${}^{155}_{64}Gd + n \rightarrow {}^{153}_{63}Eu + 2n + p - 14{,}063 MэB$$

$${}^{155}_{64}Gd + n \rightarrow {}^{153}_{64}Gd + 3n - 15{,}329 MэB$$

$$\boxed{{}^{157}_{64}Gd + n \rightarrow {}^{158}_{64}Gd + \gamma + 7{,}937 MэB}$$

$${}^{157}_{64}Gd + n \rightarrow {}^{157}_{63}Eu + p$$

$${}^{157}_{64}Gd + n \rightarrow {}^{154}_{62}Sm + {}^{4}_{2}He$$

$${}^{157}_{64}Gd + n \rightarrow {}^{153}_{62}Sm + n + {}^{4}_{2}He - 0{,}690 MэB$$

$${}^{157}_{64}Gd + n \rightarrow {}^{156}_{64}Gd + 2n - 6{,}360 MэB$$

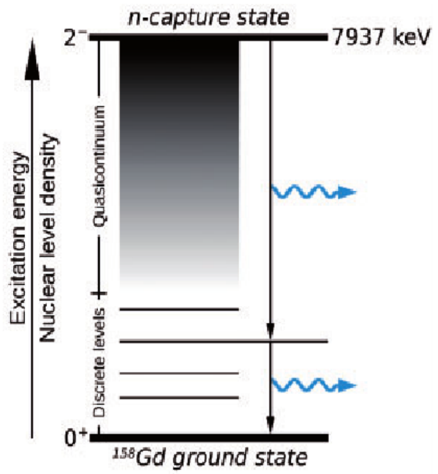
$${}^{157}_{64}Gd + n \rightarrow {}^{152}_{62}Sm + 2n + {}^{4}_{2}He - 6{,}557 MэB$$

$${}^{157}_{64}Gd + n \rightarrow {}^{156}_{63}Eu + n + p - 8{,}029 MэB$$

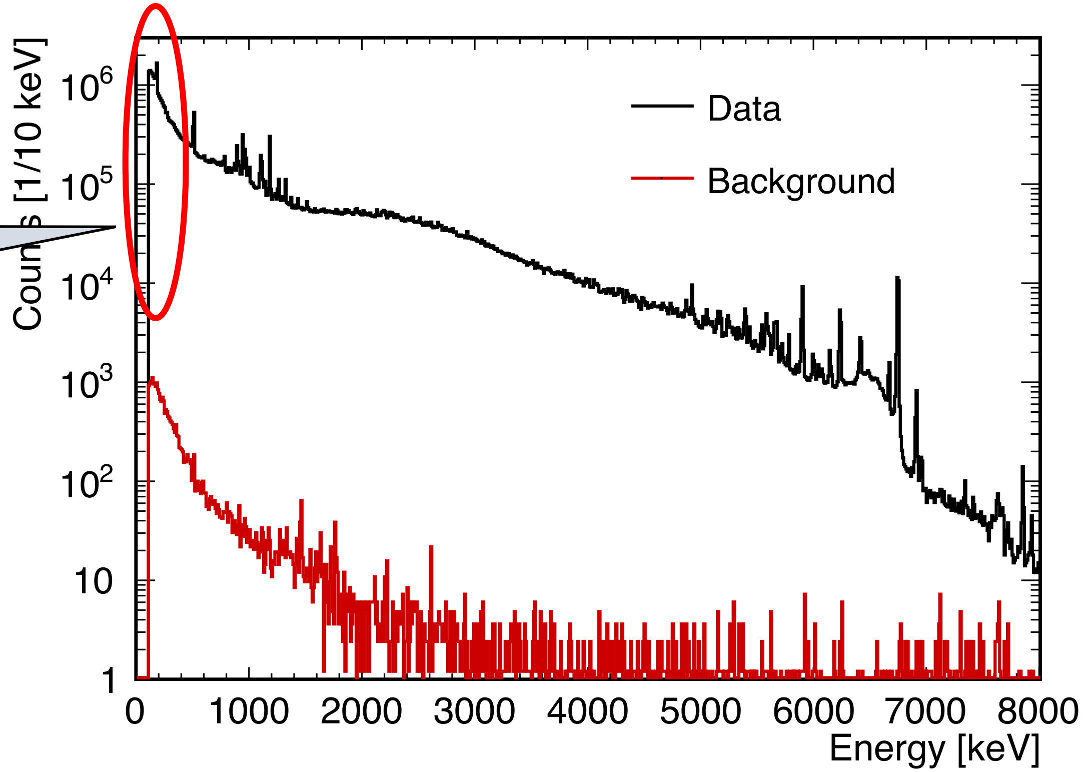
$${}^{157}_{64}Gd + n \rightarrow {}^{155}_{63}Eu + 2n + p - 14{,}366 MэB$$

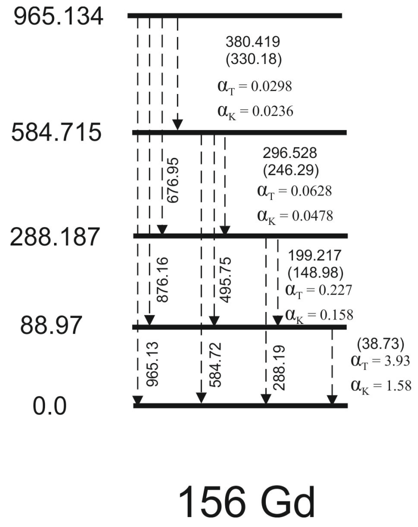
$${}^{157}_{64}Gd + n \rightarrow {}^{155}_{64}Gd + 3n - 14{,}896 MэB$$

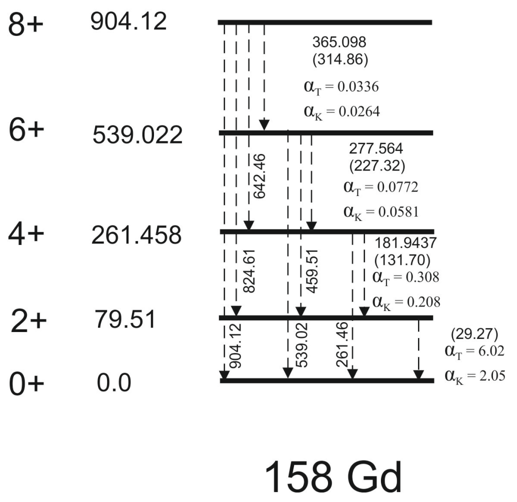
Gd nucleus relaxation through γ-quanta emission



The energy range of interest





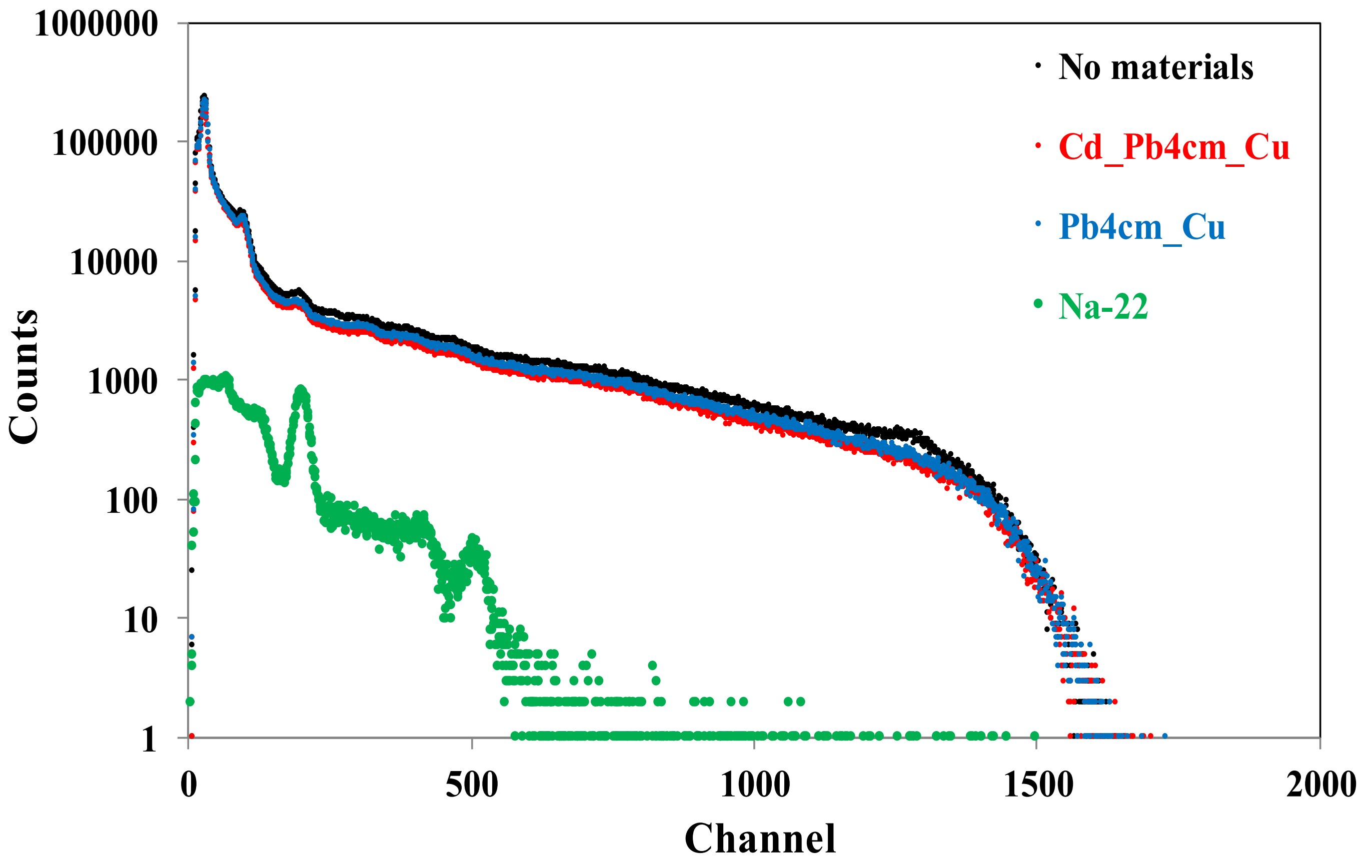


Energy spectra for events observed in a comparison with the background with the enriched 157Gd sample (black; before background subtraction).

(Kaito Hagiawara et al., Gamma Ray Spectrum from Thermal Neutron Capture on Gadolinium-157, Porg. Theor. Exp. Phys. 2015, arXiv:1809.02664v1 [nucl-ex] 10 Sept 2018, P.27)

Soft γ-quanta spectra created under neutrons of Am-Be source in GAGG and detected with the same scintillator

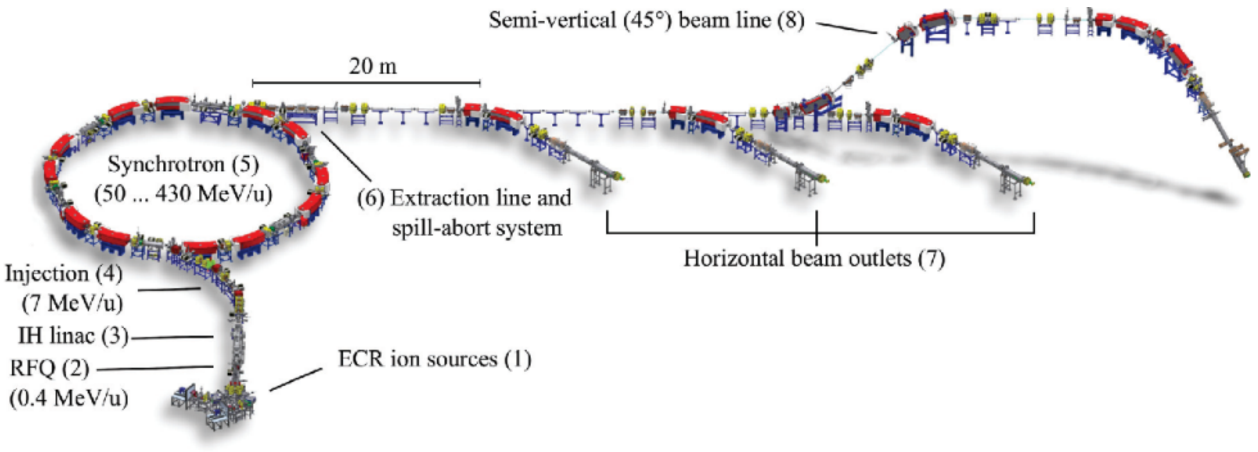
The γ- quanta spectra induced by neutrons and measured with GAGG:Ce Ø30×2 mm^3 sample.



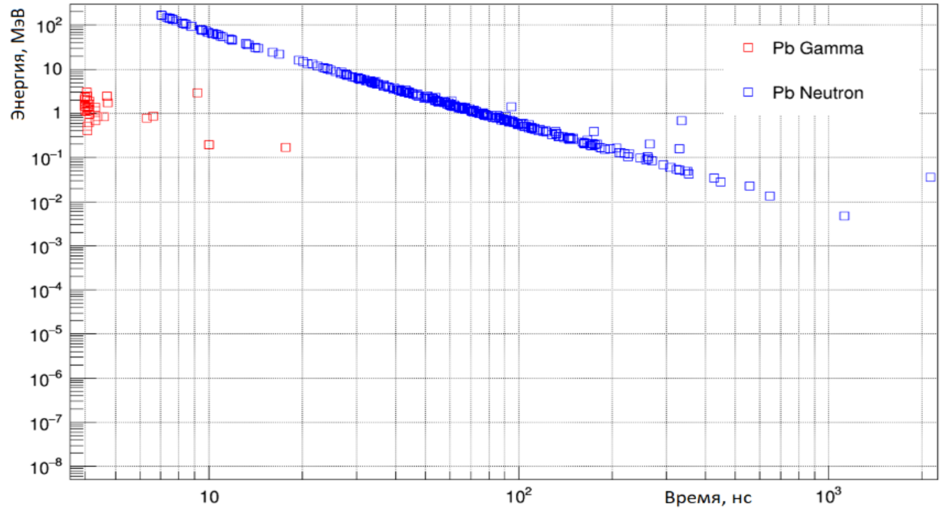
Neutron radiative capture cross section σ_γ and radiative capture resonance integral I_γ

Element	σ_γ, b	I_γ, b
Gd	49000	390
Ga	2.9	18.7
Ce	0.63	3.0
O	2.7E-4	3.1E-4
Al	0.23	0.17

TOF with GAGG scintillator

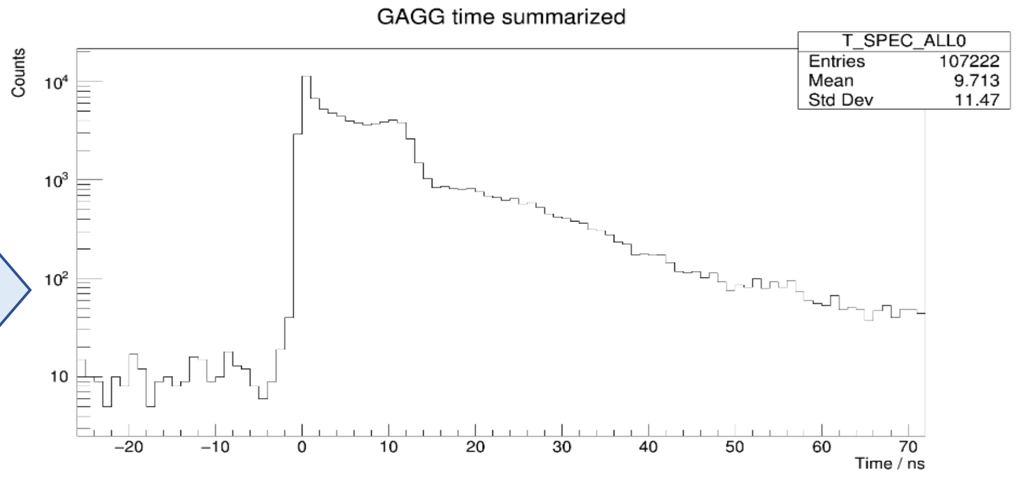


Scheme of the medical200 MeV p accelerator in Margurg (Germany)

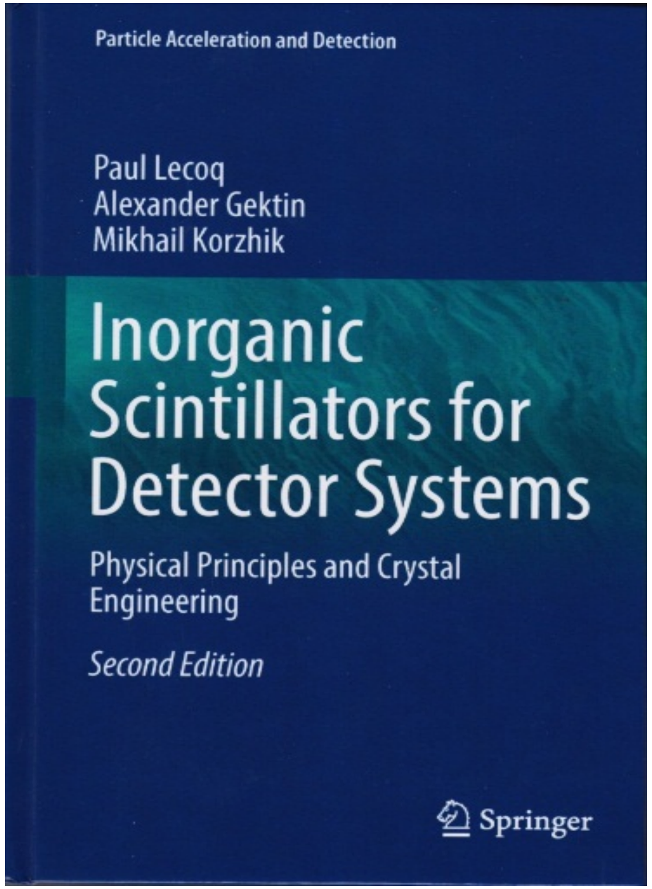


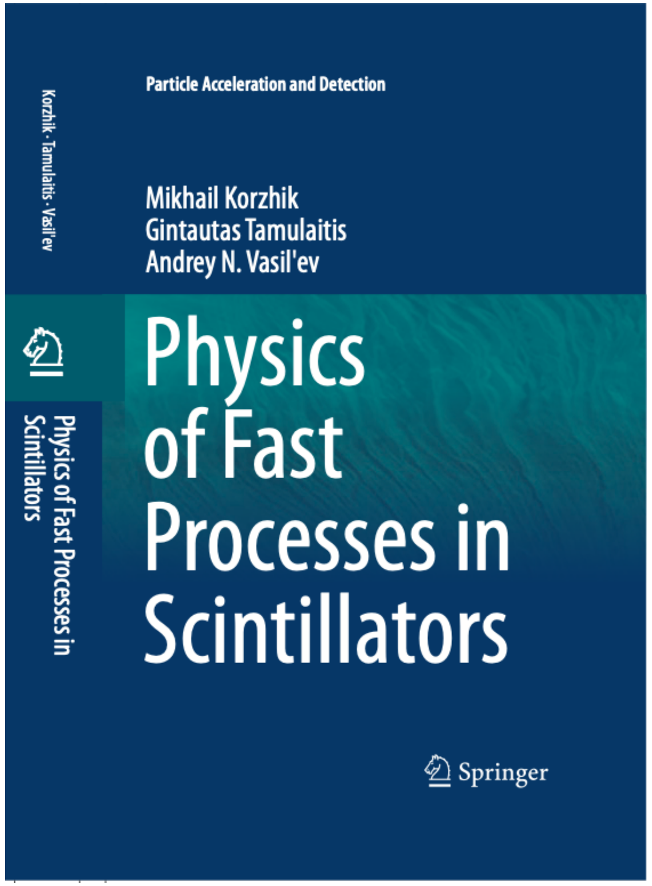
GEANT4 simulation of the neutron distribution over the time of flight from the Pb target

TOF neutron signal measured with GAGG detector. The time of -26 ns corresponds to the triggering of the start plastic counter during the passage of the proton beam pulse.



If you would like to feel the new trends in the development of the inorganic scintillators, pay attention on :





Thank you for attention and patience!