Новые сцинтилляционные кристаллы на основе кристаллов сложных оксидов

<u>О. Сидлецкий</u>, Б. Гринев, Я. Герасимов
Институт сцинтилляционных материалов НАНУ,
Украина, Харьков, пр. Ленина, 60

А. Артиков, В. Баранов, Ю. Будагов, В. Глаголев,
Ю. Давыдов, В. Терещенко
Объединенный институт ядерных исследований,
Россия, Дубна, ул. Жолио-Кюри, 6

С. Мишетти
Laboratori Nazionali di Frascati, Via E. Fermi, 40, 00044 Frascati (Roma), Italy,

Д. Хитлин
California Institute of Technology, 1200 California Blvd.,Pasadena, CA 91125, USA

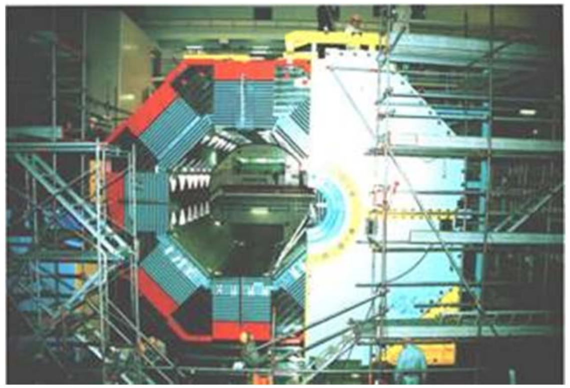
e-mail: sidletskiy@isma.kharkov.ua

Applications

- ❑ **Medical imaging**
- ❑ **Security systems**
- ❑ **High-energy physics**
- ❑ **Well-logging**

SPECT

PET



Oxide scintillators for HEP

Crystal	Experiment
BGO	**L3** **BELLE**
PWO	**CMS** **ALICE** **PANDA**
L(Y)SO:Ce	**SuperB** **Mu2e**

Scintillation characteristics of some Ce-doped oxides

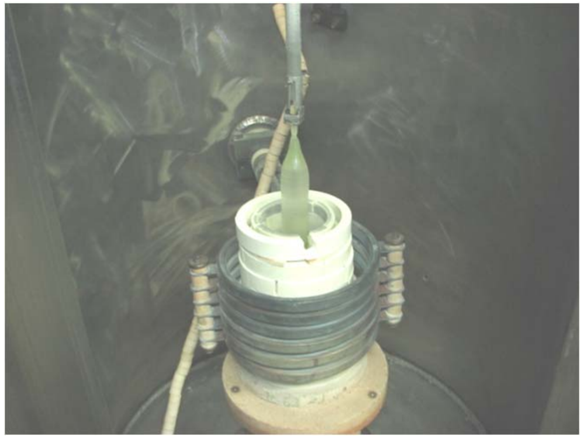
Crystal	Density, g/cm3	Light yield, phot/MeV	Energy resolution, % (^{137}Cs, 662 KeV)	Decay time, ns (γ-exc.)	Afterglow, % (after 5 ms),
Gd_2SiO_5 (GSO)	6.7	8000	9 – 11	50	0.02
Lu_2SiO_5 (LSO)	7.4	25000	7.3 – 9.7	40	> 1
$Lu_2Si_2O_7$ (LPS)	6.2	26000	9.5	38	~0.02
$Y_3Al_5O_{12}$ (YAG)	4.55	24000	7.3	85 + slow	ND
$Lu_3Al_5O_{12}$ (LuAG)	6.7	12500	ND	44	ND
$YAlO_3$ (YAP)	5.35	21000	6.7	27	ND
$LuAlO_3$ (LuAP)	8.34	11000	14	16 + slow	ND

Motivation

- ✓ Need for dense, faster and brighter scintillators for PET, security scanning systems, HEP, etc.;
- ✓ Slow decay (>300 ns) in halide scintillators (CsI(Na), NaI(Tl)…) and oxides with intrinsic luminescence ($Bi_4Ge_3O_{12}$, $CdWO_4$…). High cost of Lu- containing crystals (LSO, LYSO, LuAG).
- ✓ The required properties can be combined in mixed crystals Ce^{3+} or Pr^{3+} doped complex oxides with fast 5d-4f luminescence.

Growth facilities

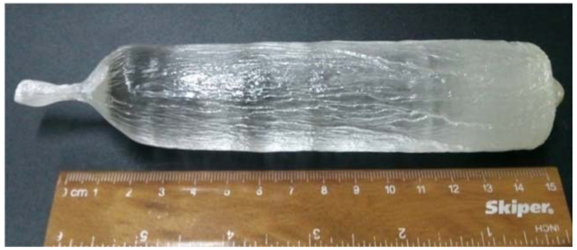
- Induction heating setups of “Oksid” and “Kristall” series for growth of crystals by the Czochralski method



- Induction heating;
- Ir crucibles
- Controlled vaccum chamber
- Diameter control by weight sensor
- Crystallization temperatures – up to ~2200°C;

Oxide scintillation crystals produced by ISMA

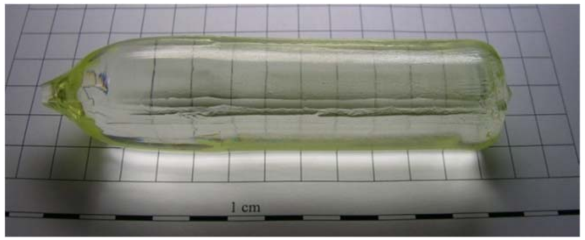
BGO up to 3” in dia.



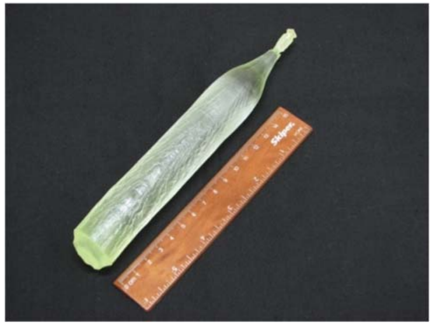
LGSO:Ce

GSO:Ce up to 2” in dia.

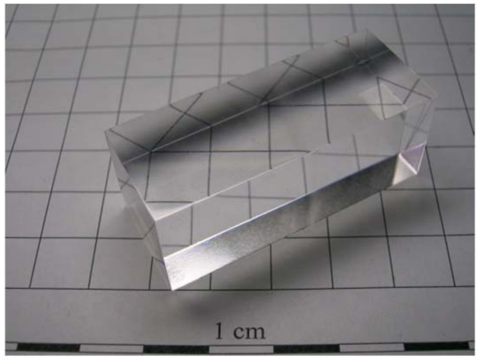
LYSO:Ce



YAG

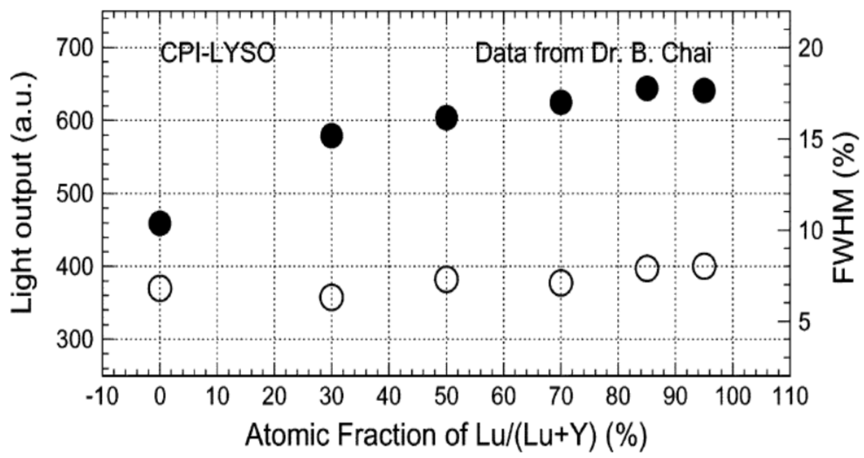


LuAG:Ce



LGSO:Ce element

$Lu_{2x}Y_{2-2x}SiO_5$:Ce (LYSO)



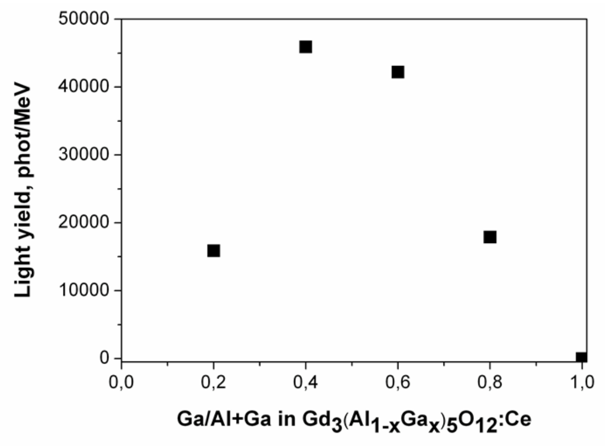
The light output (solid dots, left scale) and the energy resolution (open dots, right scale) as a function of the lutetium fraction in LYSO

[J. Chen et al.IEEE Trans. Nucl. Sci., 52, 2005) 3133]

Tm (LSO) = 2150 °C
Tm (LYSO) = ~2000-2050 °C

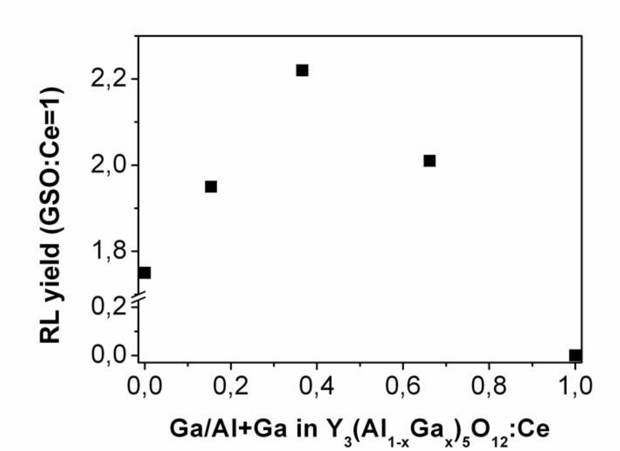
$Gd_{2x}Y_{2-2x}SiO_5$:Ce – improvement of mechanical properties compared to GSO:Ce at 0.8<x<1
[V. Bondar et al. Proc. of SCINT2005]

Light yield vs. Ga fraction in Al-Ga substituted garnets



$Gd_3(Al_xGa_{1-x})_5O_{12}$:Ce (GAGG)

Kei Kamada, et al / Cryst. Growth Des. 11 (2011), 4484–4490.



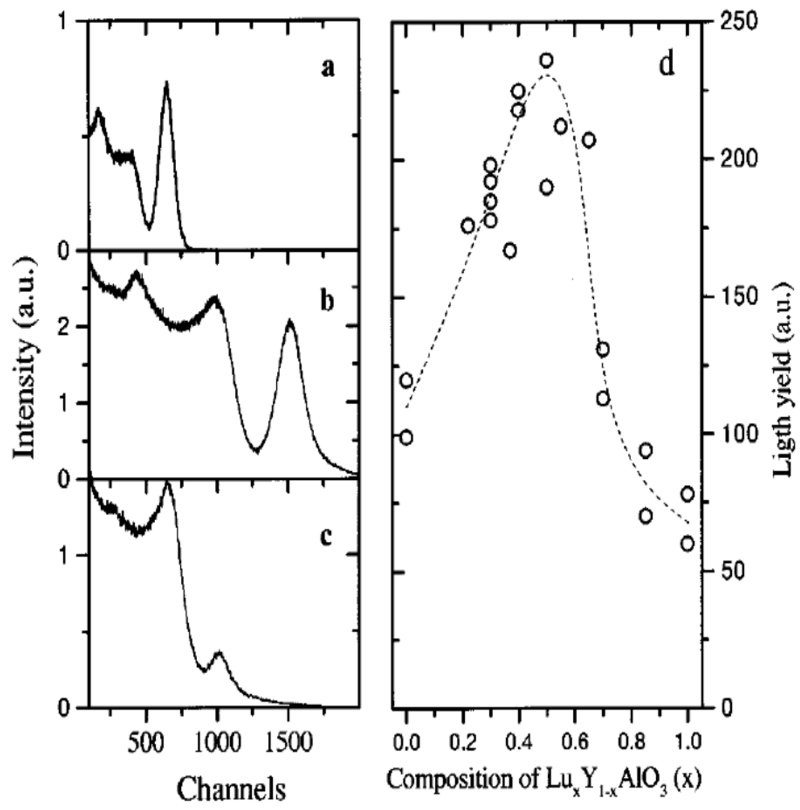
$Y_3(Al_xGa_{1-x})_5O_{12}$:Ce (YAGG)

O. Sidletskiy, et al / Materials Research Bulletin 47 (2012) 3249–3252

$Lu_xY_{1-x}AlO_3$:Ce (LuYAP)

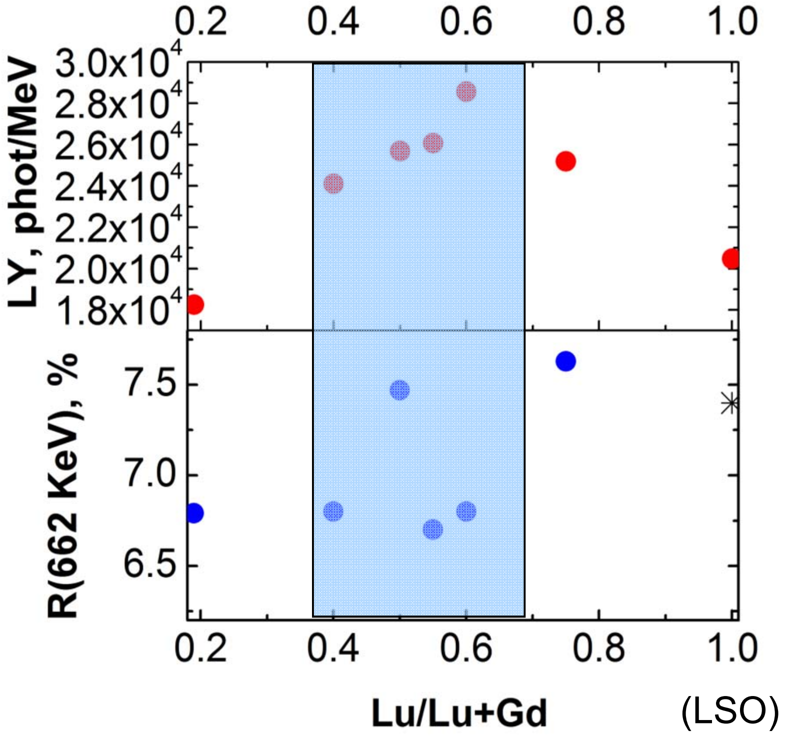
Amplitude distribution of scintillation pulses under Cs X-ray excitation:

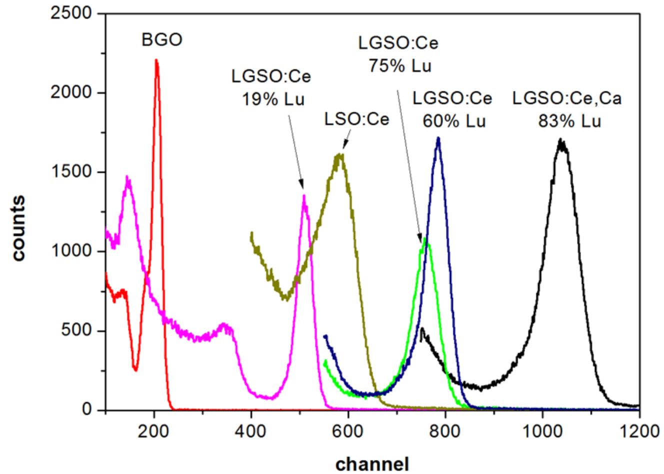
(a) LuAP:Ce,
(b) (b) LuYAP(70%Lu):Ce,
(c) YAP:Ce.
(d) Light yield measured in Ce-doped Lu Y AlO crystals of various composition.



A.N. Belsky et al. IEEE Trans. Nucl. Sci. 48 (2001) 1095

Scintillation characteristics of LGSO:Ce crystals





In the optimal host composition range (30-70 % Lu)

- ❑ Light yield 24000 – 29000 phot/MeV; (33700 phot/MeV in Ca codoped crystal);
- ❑ Energy resolution (662 KeV) - 6.7 – 7.3 %

O. Sidletskiy, V. Bondar, B. Grinyov, et al. J. Cryst Growth, 312 (2010) 601
O.Sidletskiy, A. Belsky, A. Gektin, et al. Crystal Growth & Design, (2012), dx.doi.org/10.1021/cg300608t

Bandgap engineering in rare-earth garnets $RE_3(Al_xGa_{1-x})_5O_{12}$:Ce

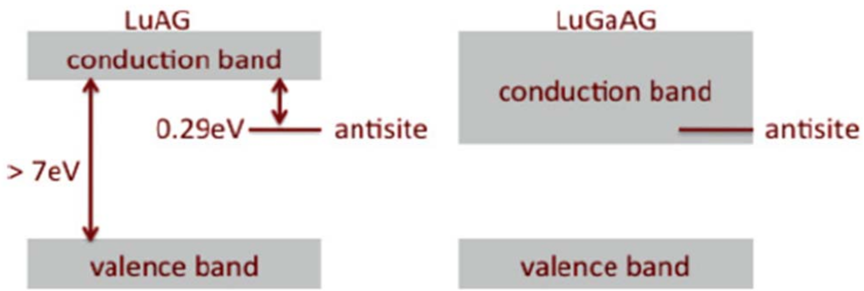
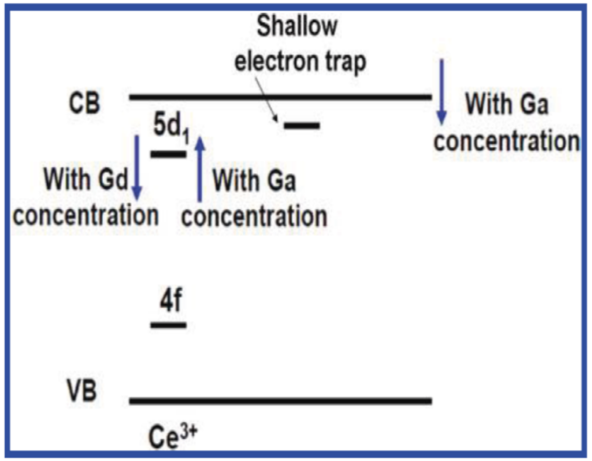


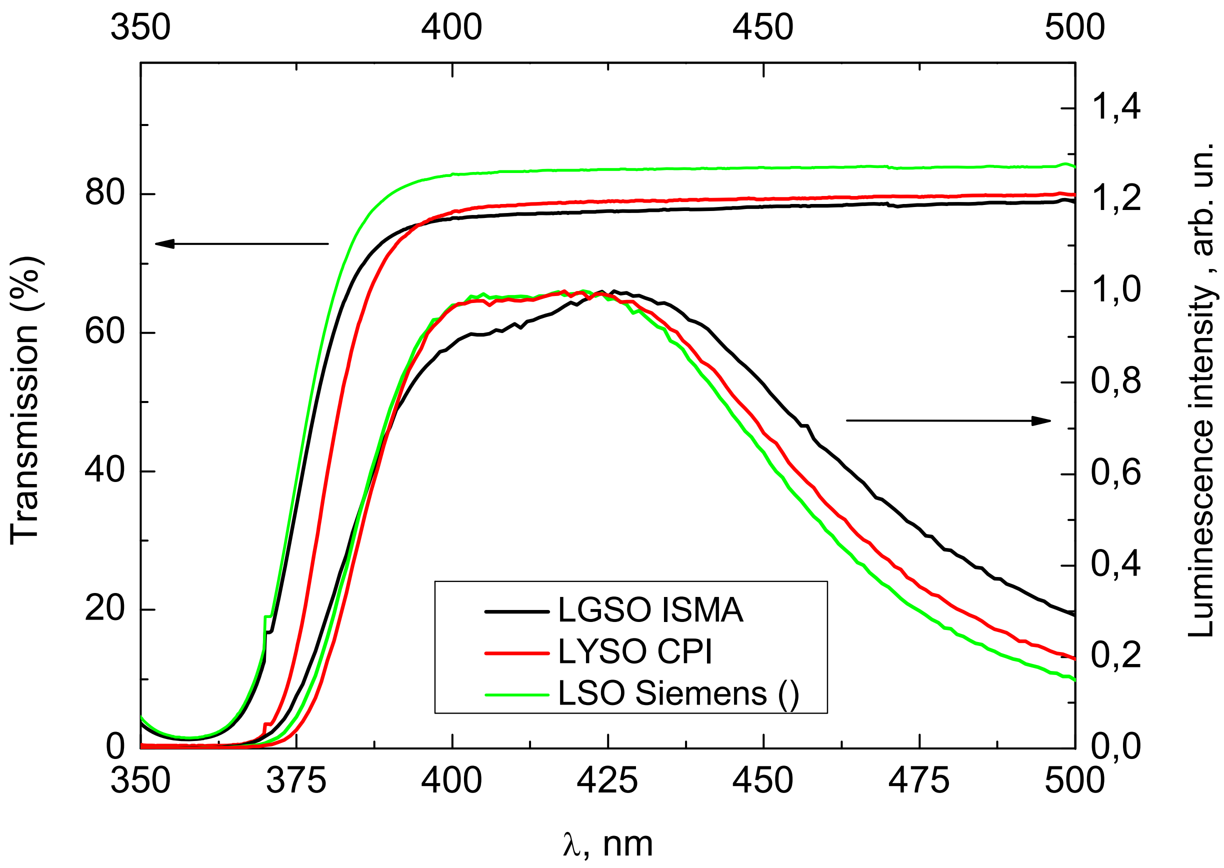
FIG. 1. (Color online) Schematic of the band structure of undoped LuAG (left-hand side), with a band gap of >7 eV and an antisite trap depth of 0.29 eV, compared to the proposed band shift due to Ga doping (right-hand side), where the antisite defect is no longer in the forbidden gap, but rather is enveloped by the CB.



Energy level scheme related to the material design

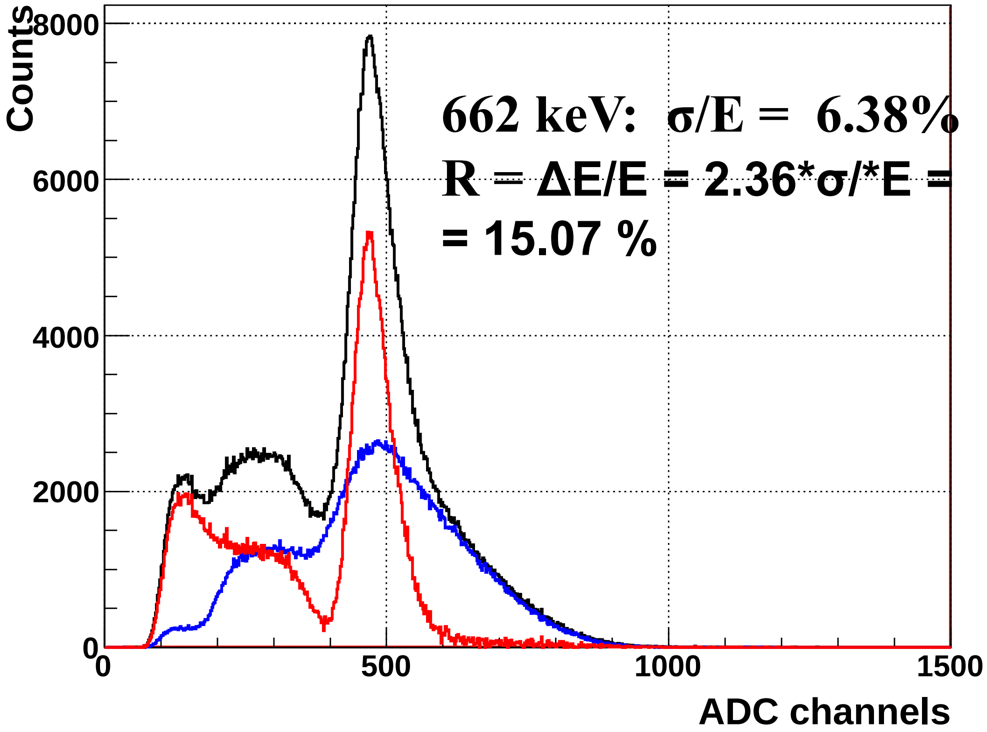
M. Fasoli et al. Phys. Rev. B 84, 081102(R) (2011)

LGSO (90 % Lu) - Transmission and X-ray luminescence



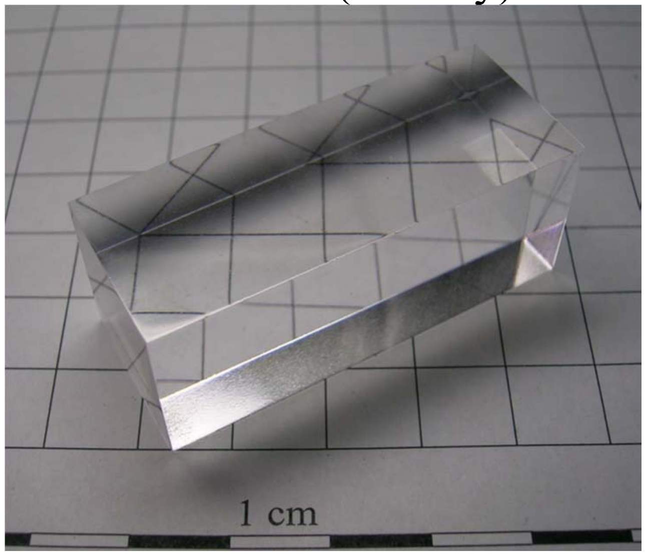
LGSO (90 % Lu) irradiation with ^{137}Cs

LGSO + ^{137}Cs rate: ~ 10480 c^{-1}
LGSO intrinsic rate: ~ 5650 c^{-1} (53.9%)

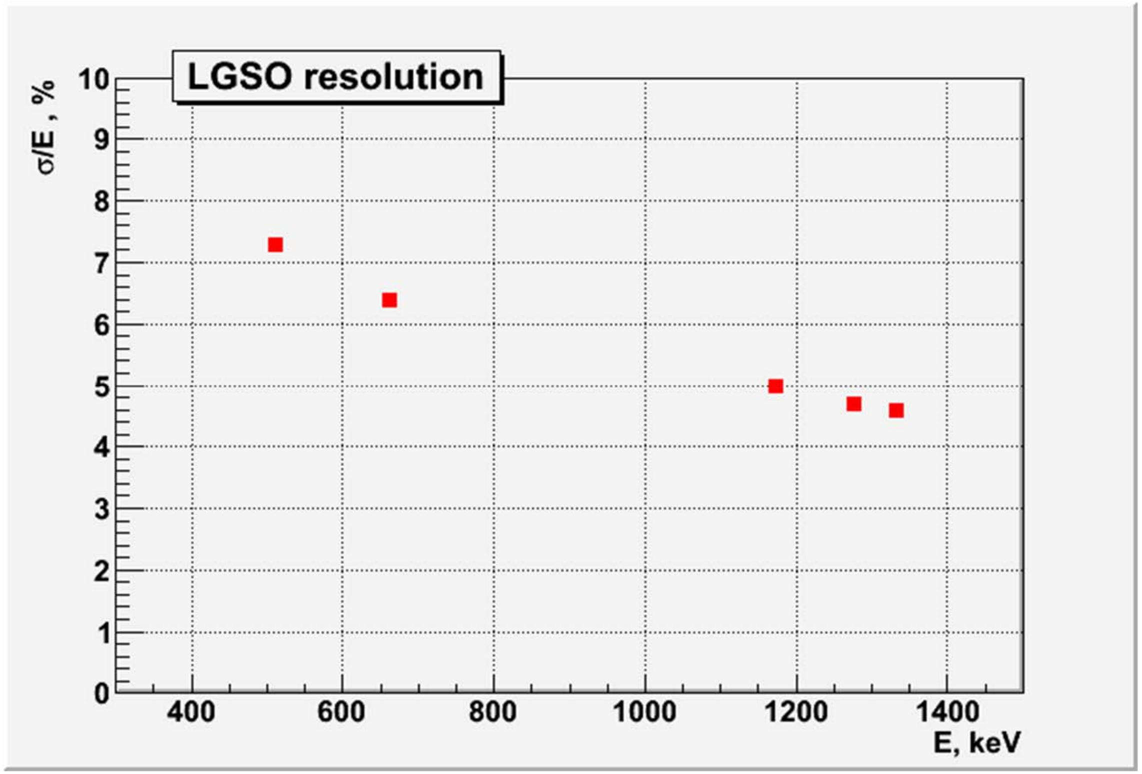


All measurements done with:

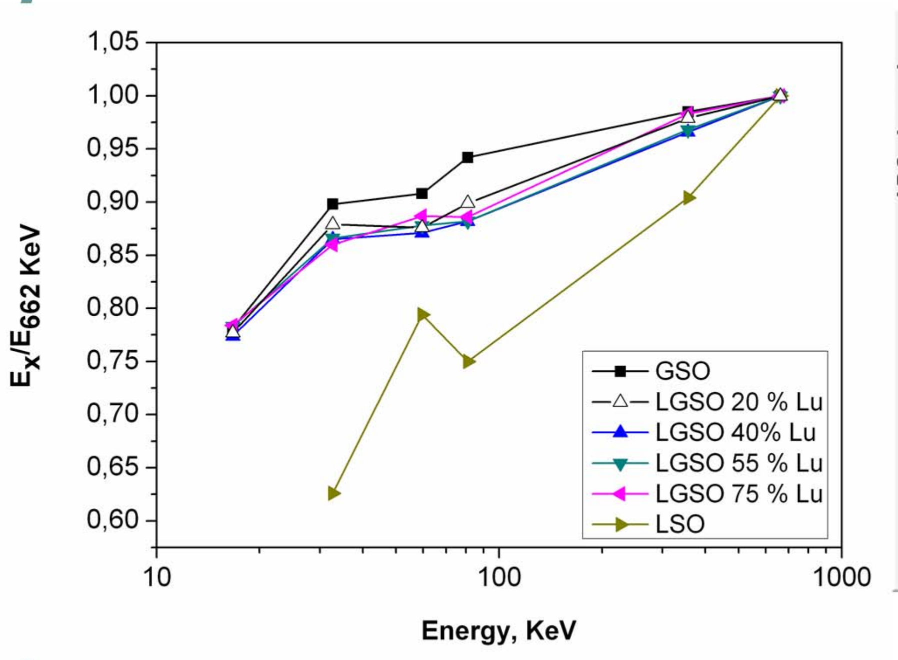
- LGSO crystal with dimensions 20x20x50mm^3
- EMI9813 PMT
- ADC 2249W (LeCroy)

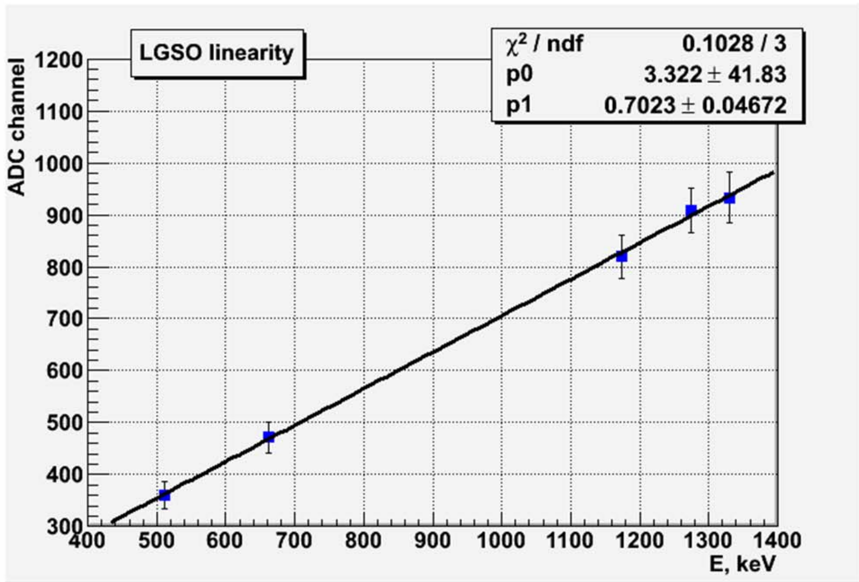


LGSO energy resolution due to gamma irradiations



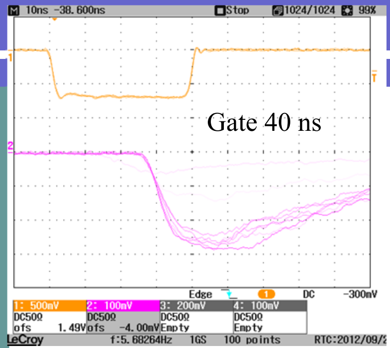
LGSO linearity

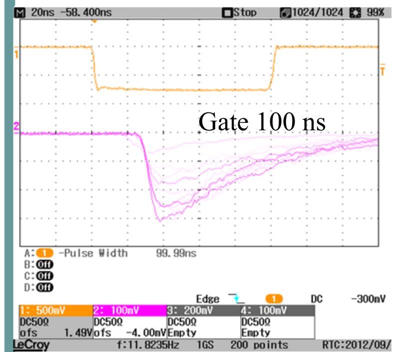


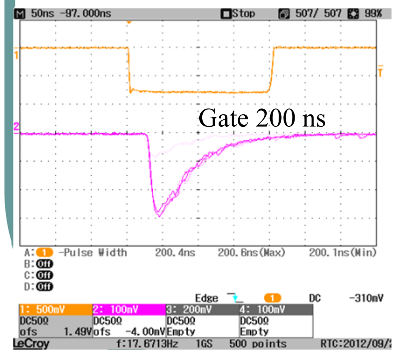


LGSO:90% Lu

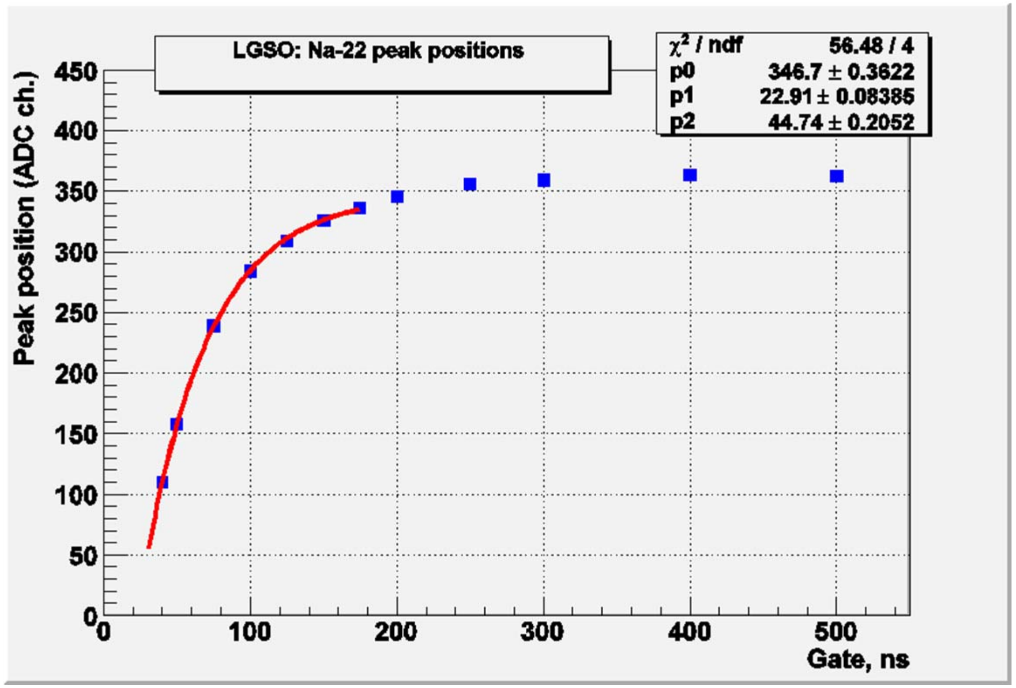
LGSO decay





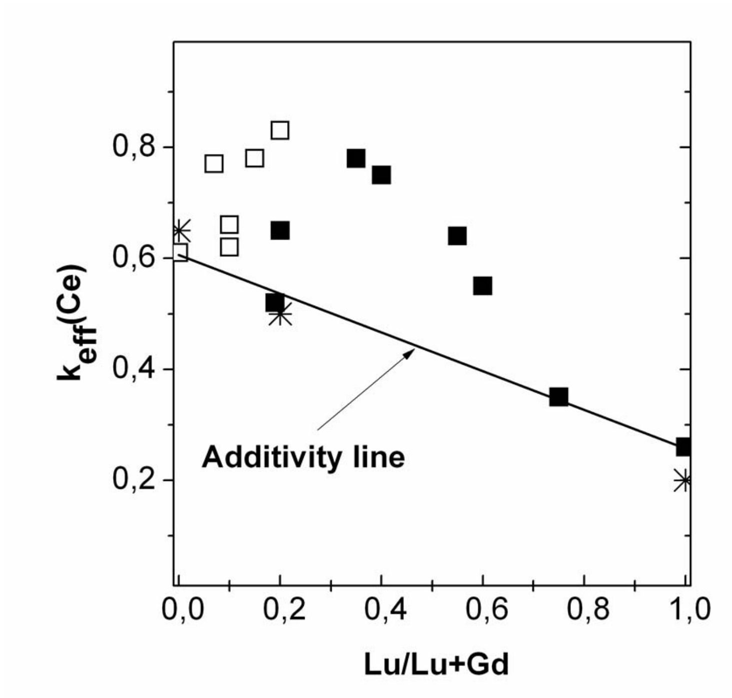


- ADC gate width varied from 40 ns to 500 ns
- ^{22}Na peak positions measured for each gate width
- Fit gives a fast component decay time ~ 44.7 ns

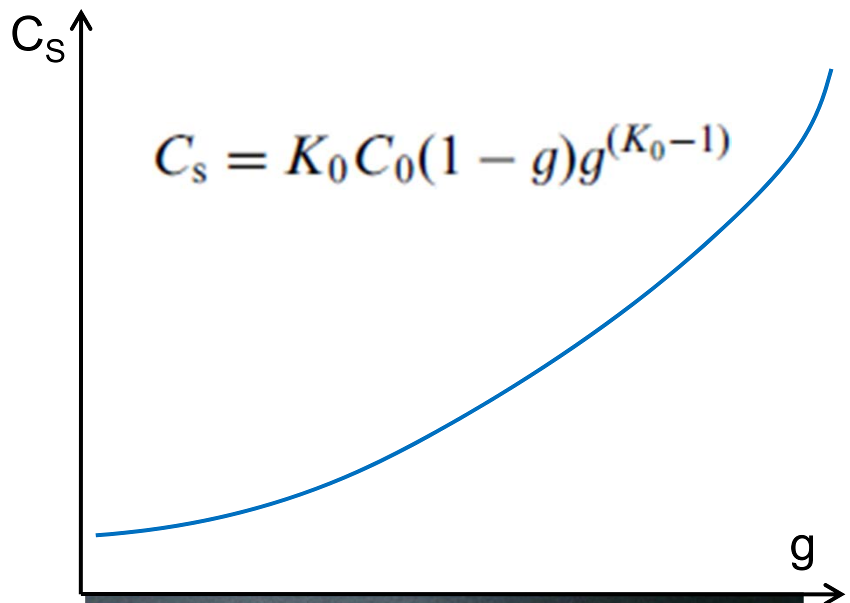


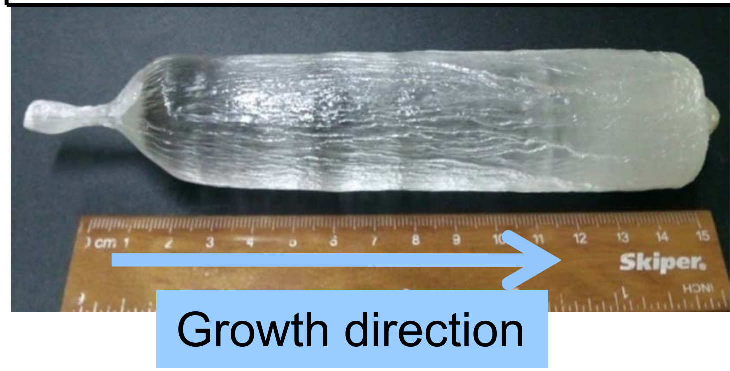
$$A = A_0 \{1 - \exp((t_0 - t)/\tau)\}$$

Distribution coefficient of Ce



Non-additive increase of k_{eff} (Ce) in LGSO:Ce





Выводы

- Оксидные сцинтилляторы широко используются в экспериментах физики высоких энергий благодаря высокой плотности и Z_{eff}, негигроскопичности, быстрому затуханию, хорошей радиационной стойкости.

- Недостатками оксидных сцинтилляторов является высокая стоимость (для высокотемпературных кристаллов) и относительно невысокий световой выход

- Недавно были разработаны новые смешанные кристаллы с улучшенными сцинтилляционными параметрами:
  - LGSO:Ce – СВ до 34000 фот/МэВ, R(662 KeV) = 6.4 %;
  - GAGG:Ce – СВ до 65000 фот/МэВ, R(662 KeV) = 4.6 %;
  - YAGG:Ce – СВ до 25000 фот/МэВ;