Lawrence Livermore National Laboratory

Scintillator Materials for Gamma Ray Spectroscopy

February 4, 2011

Presented at RIKEN Nishina Ctr., Wako, Japan
as part of the
"SHOGUN Gamma Ray Spectrometer Symposium"

Funded by DHS/DNDO and DOE NA-22

N.J. Cherepy, cherepy1@llnl.gov
S.A. Payne, B.W. Sturm, J.D. Kuntz, Z.M. Seeley, B.L. Rupert, R.D. Sanner,
T.A. Hurst, P. Thelin, S.E. Fisher, O.B. Drury
Lawrence Livermore National Laboratory, Livermore, CA 94550
K.S. Shah and team, *Radiation Monitoring Devices*
A. Burger and team, *Fisk University*
L.A. Boatner and team, *Oak Ridge National Laboratory*

LLNL-PRES-468519

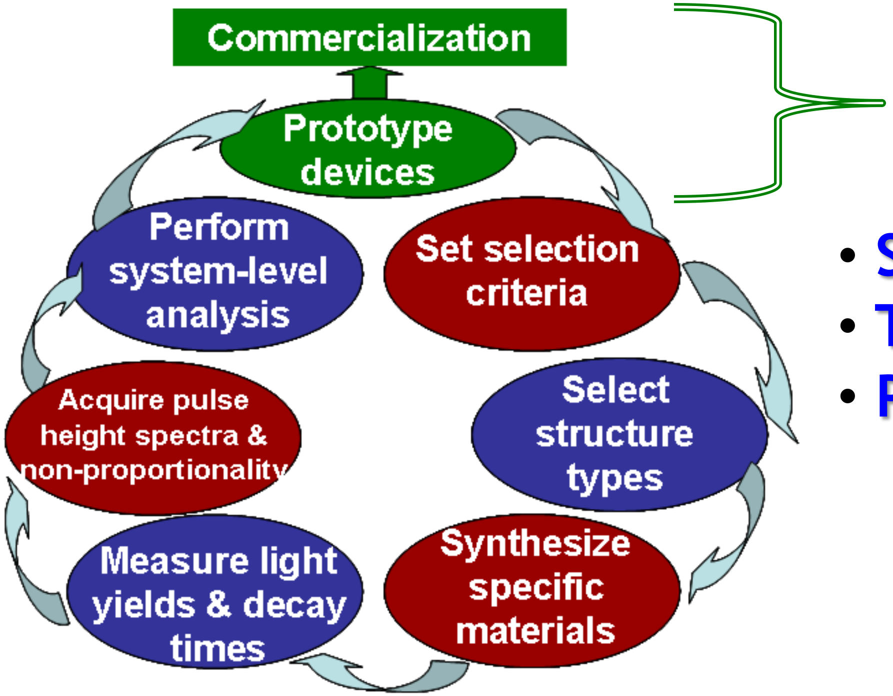
This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

Overview

Problem: Accurate measurement of Doppler-shifted gammas produced in rare isotope beams during decay, fragmentation and nuclear reactions

Requirements for new detector materials:

1) High energy resolution and stopping – to discriminate gamma spectra
2) Fast coincidence timing w/ low dead time – to observe correlated events
3) Radiation hardness – for high rate / long duration experiments
4) Low cost / maintenance – starting materials; growth / ruggedness; 1000's of units
5) Fabrication into small cuboids – to achieve close-packing and geometrical segmentation



Our **Directed Search Method** has been used to discover:

- **Single Crystals** → $SrI_2(Eu)$
- **Transparent Ceramics** → Garnet(Ce)
- **Plastics** → Bi-loaded Polymer

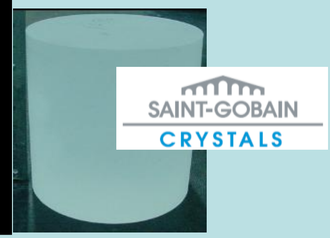
For radioisotope identification, gamma scintillators with high resolution, low cost and large volume are needed

CURRENT

Gamma Scintillation Detectors

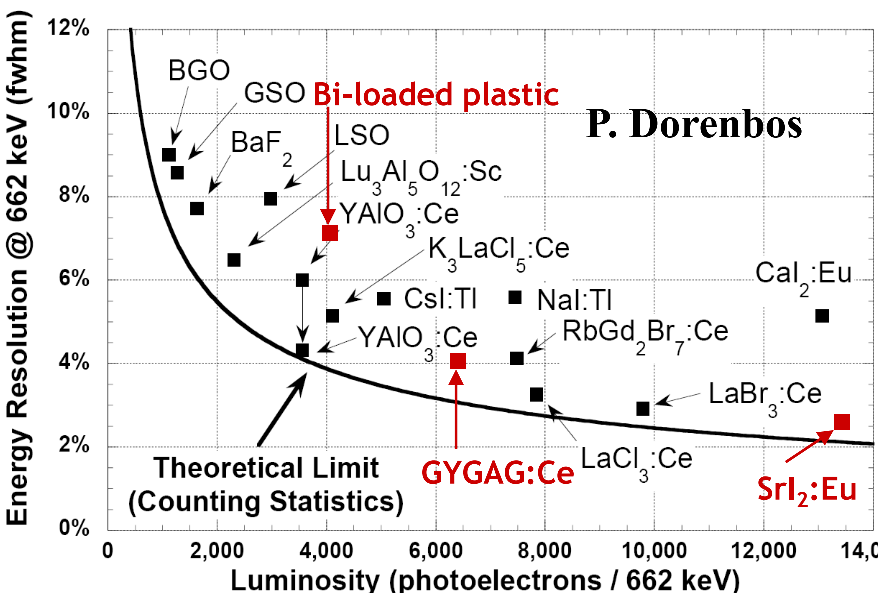
NaI(Tl)
- 6% resolution
- Room temperature
- 2000 cm^3
- \$10/cm^3

LaX$_3$(Ce)
- 3% resolution
- Room temperature
- 400 cm^3
- \$100/cm^3
- Self-activity



FUTURE

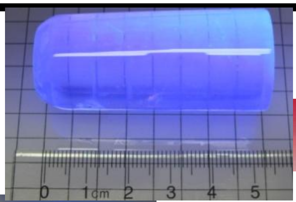
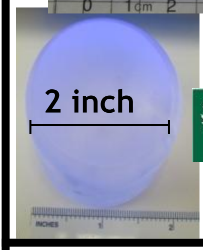
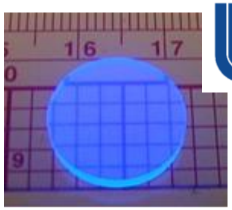
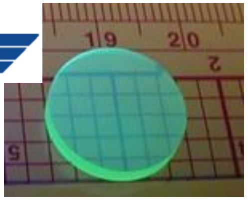
Goals:
- 2% resolution
- Room temperature
- 2000 cm^3
- \$10/cm^3
- No self-activity



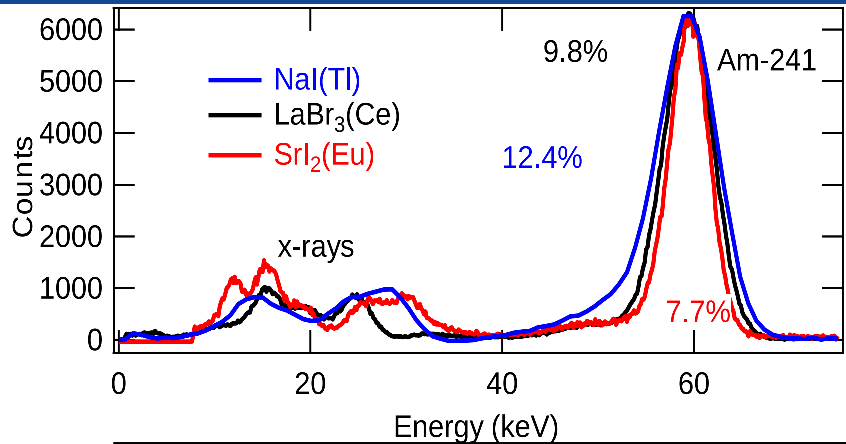
High resolution enabled by:

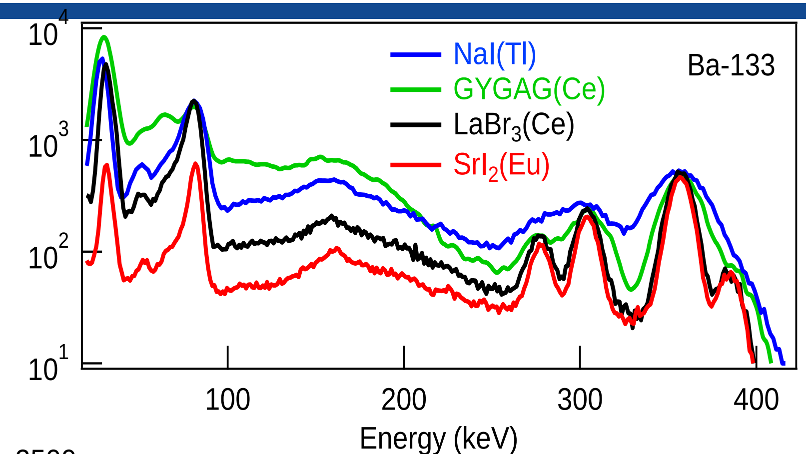
-High Light Yield
-Low Non-Proportionality
-Uniform Material Response
-Optimized Light Collection
-Accurate Readout

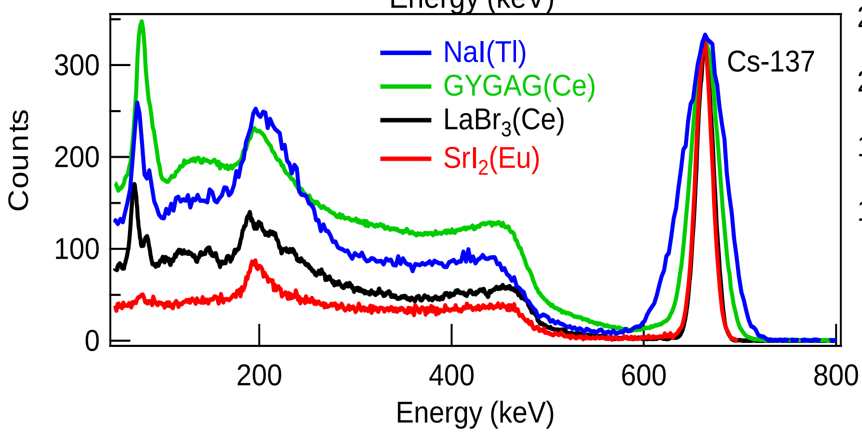
Inorganic single crystal and ceramic scintillators are being developed for gamma ray spectroscopy

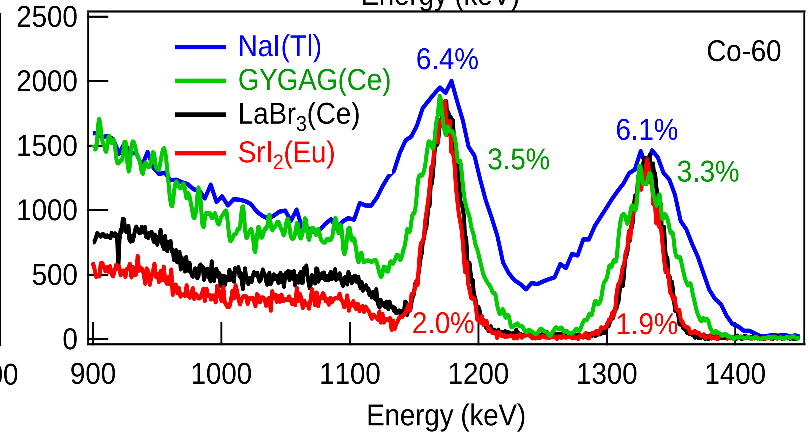
Single crystals	Ceramics	Plastics
$SrI_2(Eu)$   	GYGAG(Ce), 1 in^3 	  Bi-loaded polymers, 1 cm^3
▪ Often hygroscopic/air-sensitive ▪ Fragile/brittle ▪ Complex to grow large crystals ▪ Can have gradients & non-uniformity	▪ Unreactive with air, water ▪ Mechanically durable ▪ Large sizes (100 cm^3 Nd:YAG ceramics commercially available) ▪ Increased activator uniformity ▪ Can form high melting point oxides	▪ Unreactive with air, water ▪ Mechanically durable ▪ Large sizes, low cost
▪ All crystal structures possible ▪ Best energy resolution materials- $LaBr_3(Ce)$, $SrI_2(Eu)$ ~2.6% @ 662 keV	▪ Requires cubic material ▪ Good energy resolution- GYGAG(Ce) Gadolinium Garnet ~4.5% @ 662 keV	▪ Non-standard polymer required ▪ Bi-loading uniformity important ▪ Energy resolution so far ~7% @ 662 keV

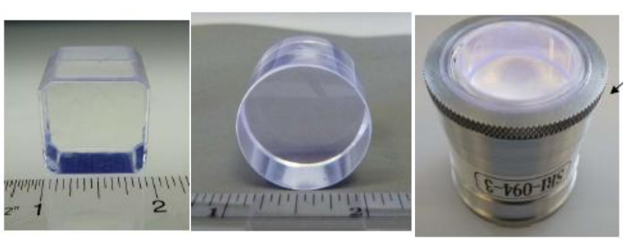
For gamma ray spectroscopy, SrI_2(Eu) comparable to $LaBr_3$(Ce) and GYGAG(Ce) is better than NaI(Tl)



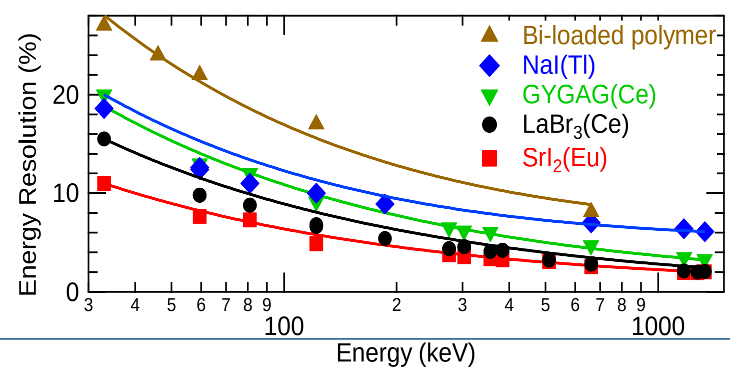




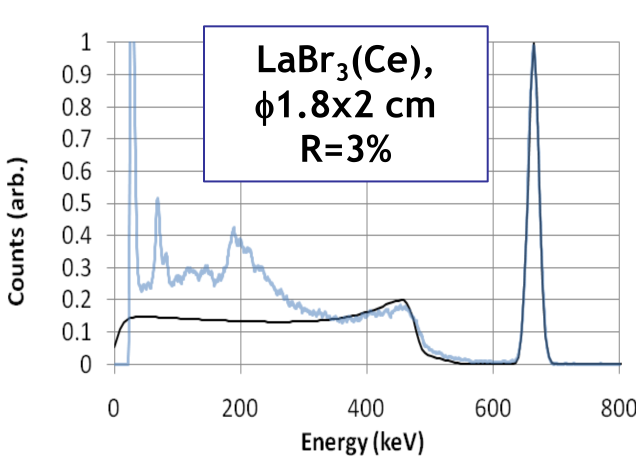


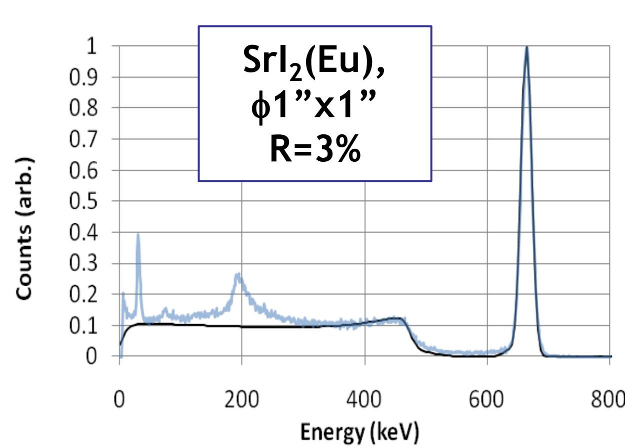


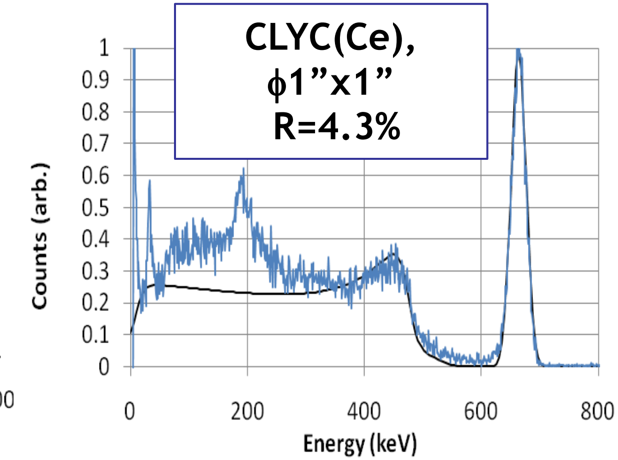
SrI_2(Eu) crystals grown at RMD, Inc.
Cut, polished and encapsulated by LLNL

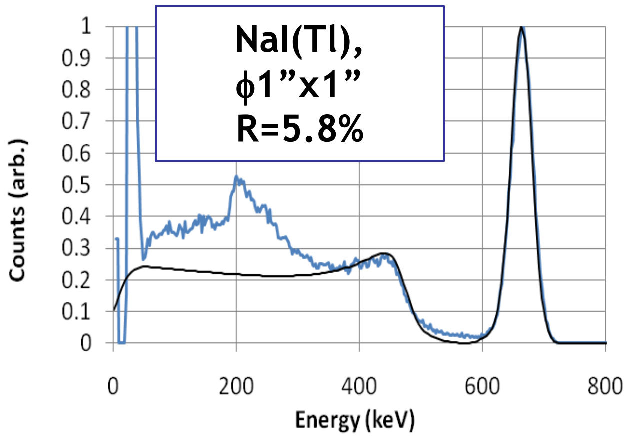


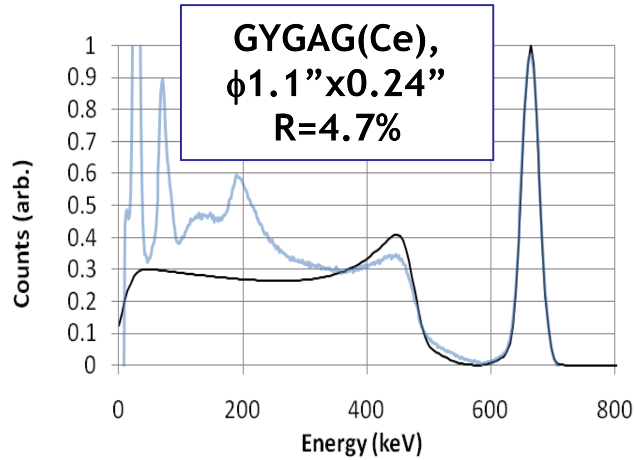
Comparison of gamma spectra at 662 keV for ~(1")^3 scintillators reveals effects of resolution and photopeak efficiency, S_{PP}









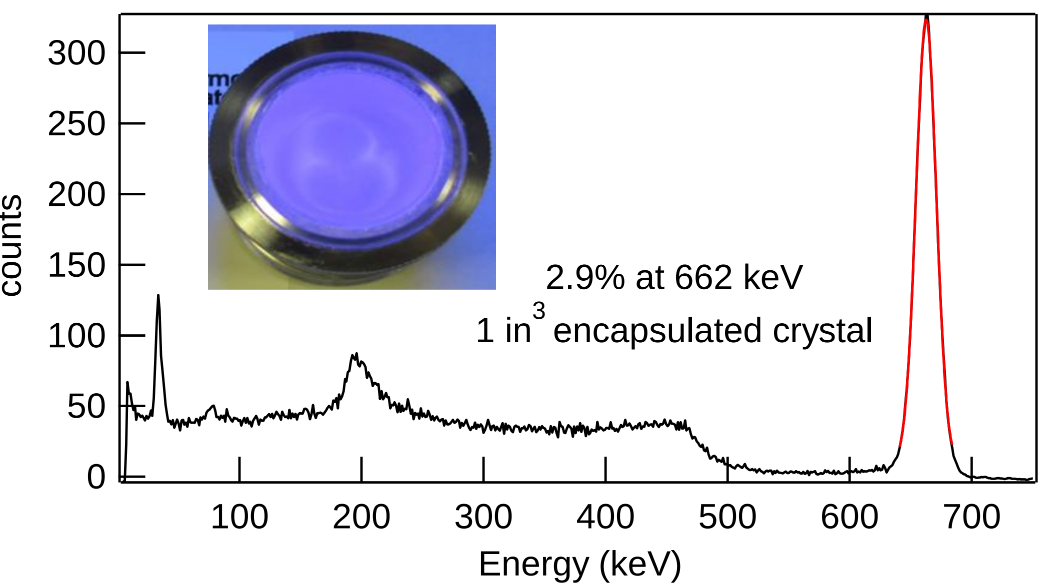


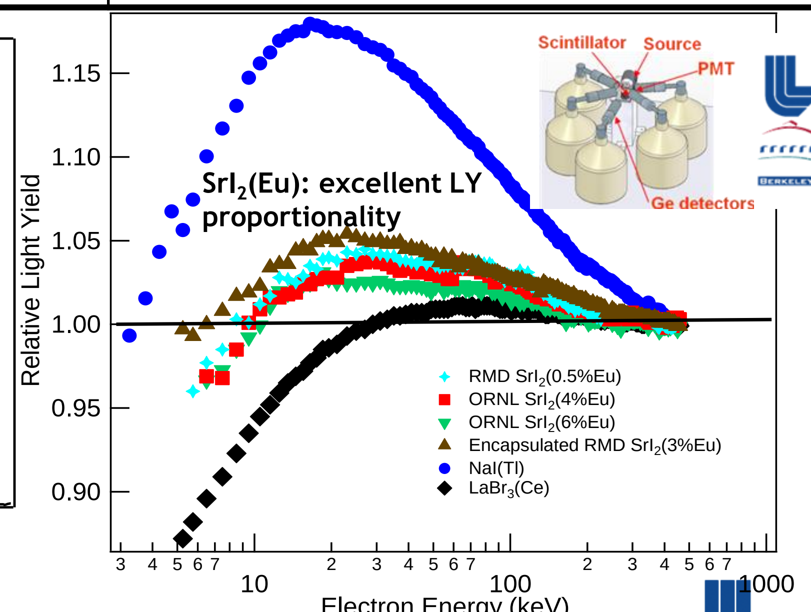
Scintillator	S_{PP} (662kev) for 1"x1" (%)
$LaBr_3$(Ce)	11.1
SrI_2(Eu)	11.4
CLYC(Ce)	5.3
NaI(Tl)	8.5
GYGAG(Ce)	12.8

- For comparison, 1"x1" Ge photopeak efficiency, S_{PP} = 3.9%
- S_{PP} defined as fraction of gammas intercepting detector that lead to photopeak

Production of encapsulated $SrI_2(Eu)$ underway

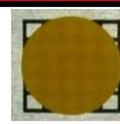
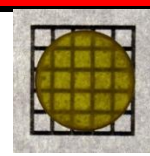
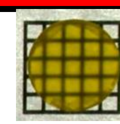
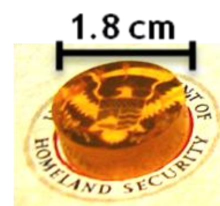
Property	$LaBr_3(Ce)$	$SrI_2(Eu)$	Comparison
Melting Point	783 ˚C	538 ˚C	✓ Less thermal stress
Handling	Easily cleaves	Resists cracking	✓ Better processing
Light Yield	60,000 Ph/MeV	90,000 Ph/MeV	✓ Higher
Proportionality contribution	~2.0%	~2.0%	✓ Favorable
Inhomogeniety	0%	>1% (current)	Impurities and surfaces being addressed
Decay time	30 nsec	0.5-1.5 μsec	Fast enough to avoid deleterious signal pile-up
Self-radioactivity	La ~ 3x NORM	None	✓ Less noise
Hygroscopic / air sensitive?	Very	Very	Similar
γ absorption (2x3", 662 keV)	22%	24%	Similar

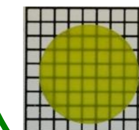




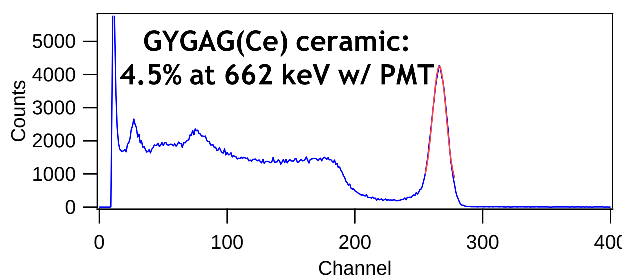
We have been working to identify an optimal Gd-based garnet scintillator for the past five years

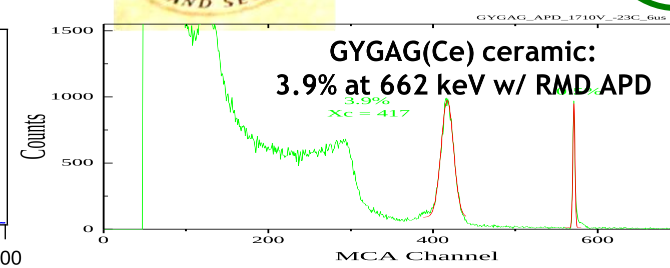
	2006-7		2007-8		2008-9		
Composition	GAG	GYAG	GGG	GSAG	GYSAG	GGAG	GYGAG
Phase Stability	Poor	Moderate	Excellent	Excellent	Excellent	Moderate	Excellent
γ-LY(Ph/MeV)	___	40,000	___	25,000	30,000	55,000	50,000
En. Res. (662 keV)	___	11%	___	11%	10%	9%	4-5%

2010-11
Scale-up of GYGAG(Ce)





Current status:

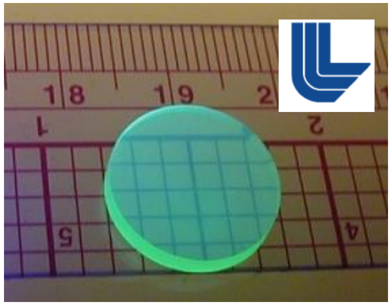
- 1 in^3 parts formed routinely

To optimize performance:

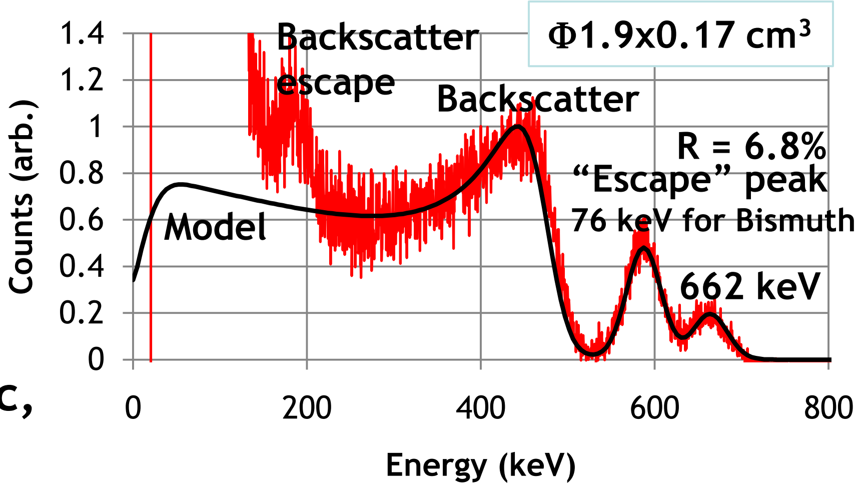
- Photodetector matched to green scintillation

Recently we have developed polymer scintillators with enhanced scintillation characteristics

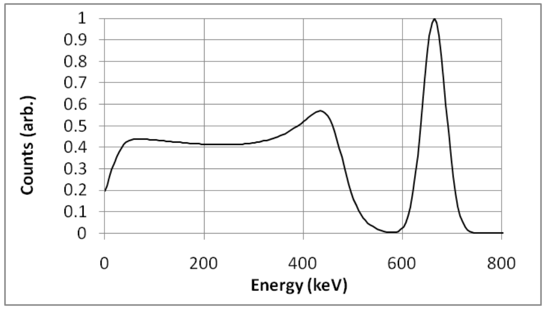
Gamma Spectroscopy Plastic: Energy resolution similar to NaI(Tl)



Bi organometallic, 40 wt%

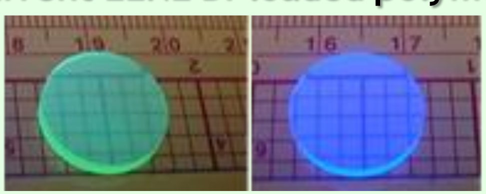


Predicted 3" part w/ R= 8%



Escape peaks in small parts eliminated when larger

Materials for future gamma spectrometers?

	Gamma Spectroscopy Scintillator	Z_{eff}	Light Yld (Ph/MeV)	En Res, 662 keV	Nonprop En Res, 662 keV	S_{PP}, 1 MeV, (15 mm x 15mm x 15 mm)
Single crystals	NaI(Tl)	50	40,000	7%	5.0%	2.2%
	$LaBr_3$(Ce)	44	63,000	3%	2.2%	2.8%
	SrI_2(Eu) 	49	90,000	3%	2.2%	3.0%
Garnet ceramics	$(Gd,Y, Lu)_3(Al,Ga)_5O_{12}$(Ce) 	47	50,000	4.5%	1.9%	3-9%
Plastics	Standard PVT	4.5	15,000	8% (Compton)	3.5%	0
	Current LLNL Bi-loaded polymers 	26	10,000-30,000	7%	3.5%	0.3%