Crystal scintillators for low background measurements

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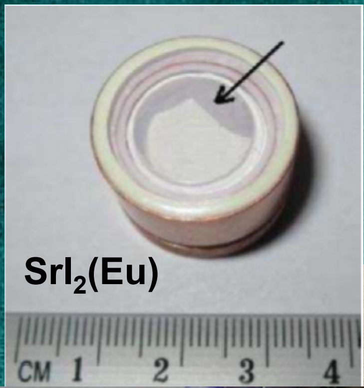
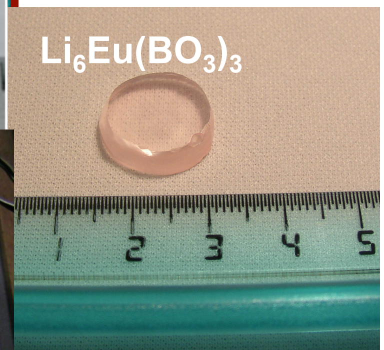
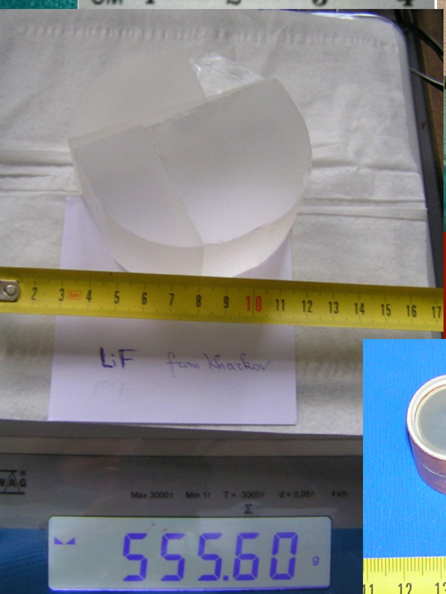
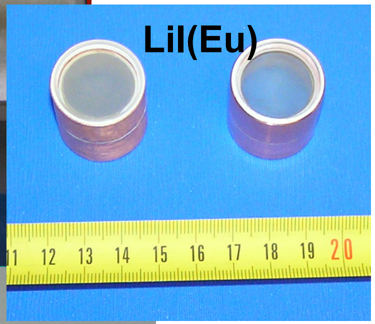
LRT2013, LNGS

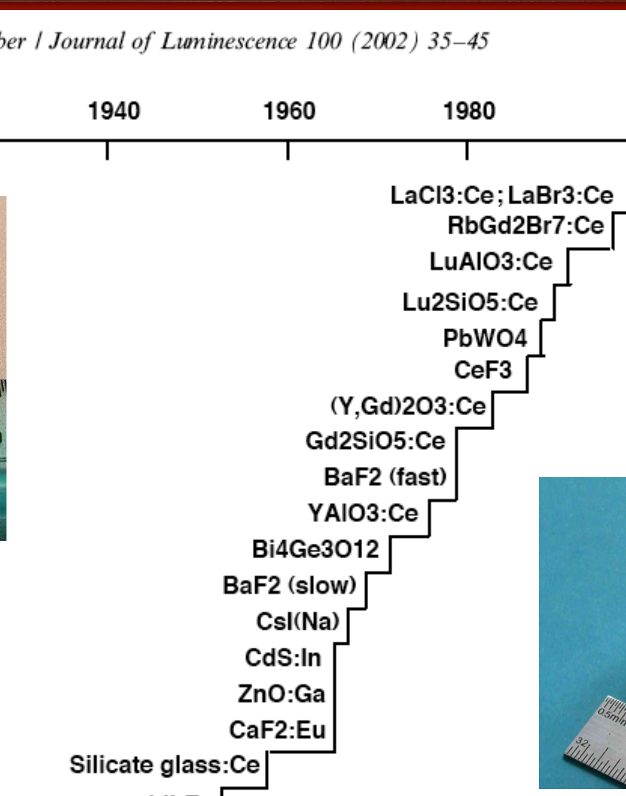
Continuing innovation

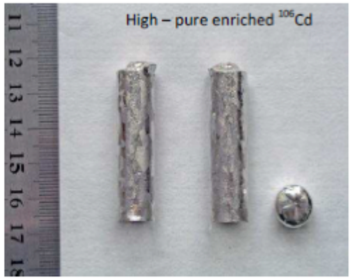


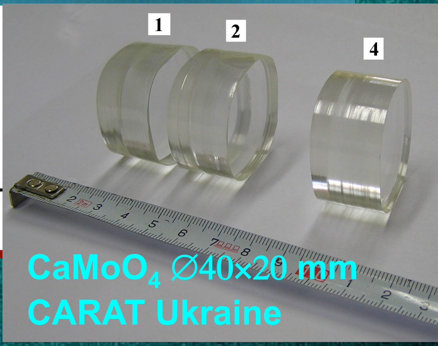












Crystal Growth: main methods

Method	Advantages	Drawbacks
Bridgman–Stockbarger	The simplest method for alkali halide crystals	Direct contact of the crystal with the crucible walls. Stresses in the growing crystal and extraction (cracks). Difficult the uniform activator distribution through the ingot. Insufficient convectional melt mixing before the crystallization front (inclusions and striations). Spontaneous crystallization on the ampoule surface (the orientation of the crystal is difficult to control), a well-oriented seed is needed.
Czochralski	No direct contact of the growing crystal with the crucible walls. Allows an increase in crystal growth rate of several times owing to a higher axial and radial temperature gradients and due to an intensive mixing of the melt by the rotating crystal. Seems to be the most efficient when crystals are required with high structural perfection.	More complicated technically; permanent control and correction of the main parameters needed. If non automated pulling the success of growth is defined mainly by the skill of the operator.
Kyropoulos	No direct contact of the growing crystal with the crucible walls. Highly controlled thermal gradient keeps a low-stress environment for the crystal.	A deep control of all the parameters needed

Implementations of Czochralski -- Kyropoulos techniques are operative and in evolution

Detector Performances

They depend on many specific parameters as e.g.:

The growth parameters (geometrical, thermal, chemical, supersaturation, hydrodynamic) and their nonlinear interrelations.
Nonstoichiometry, i.e. deviation from congruent melting composition.
Impurities often have a significant impact on the crystal-growth processes and on optical properties.
The crystal machining, **the parts used** of the ingot, **the surface preparation**, **the environment**, **the protocols** have to be deeply studied and then optimized
In addition: **purity of the components**, **purity of additives and dopants**, **time and environment of storing materials** etc.
important parameters for optical properties: form and size of the crystal, crystal surface treatment, reflecting materials, bond scintillator-photoreceiver.

+ requirements for low background

Crystal scintillators@LNGS
DAMA/R&D
rare proc
DAMA/CRYS
rare proc
DAMA/LIBRA
DM and rare proc
ββ decay modes
Dark Matter candidates
of various nature
Possible CNC processes
Matter stability
Electron stability
Rare nuclear decay modes
Search for exotics in cosmic rays
Solar axions
Etc…
DM CRESST
($CaWO_4$+$ZnWO_4$)
Support by LNGS-Chemistry Lab(ICP-MS, analysis/tests, cleaning, purifications etc)
Support by STELLA Facility on radio-purity of samples and low backgr measurements
ββ decay LUCIFER
(LUMINEU/AmoRE)
(ZnSe,$ZnMoO_4$,$CaMoO_4$)
LUCIFER
@LNGS also scintillating crystals as bolometers

Crystal scintillators in DM and ββ investigation

Experiment	Target	Type	Status	Site
ANAIS	NaI	annual modulation	construction	Canfranc
DAMA/NaI	NaI	PSD/annual modulation	concluded	LNGS
DAMA/LIBRA	NaI	annual modulation	running	LNGS
DAMA/1 ton	NaI	annual modulation	R&D	LNGS
DM-ICE	NaI		prototype test	South Pole
ELEGANT V	NaI	ββ	concluded	Kamioka
LSM	NaI	PSD	concluded	LSM
NAIAD	NaI	PSD	concluded	Boulby
PICO-LON	NaI	segmentation	->KamLAND-PICO	Oto
KIMS	CsI,(NaI)	PSD/annual modulation	running	Y2L(Korea)
TEXONO	CsI	neutrino phys.	running	KS Lab
CaF_2-kamioka	CaF_2	DM	concluded	Kamioka
CANDLES	CaF_2	ββ and DM	running	Kamioka
DAMA-CaF_2	CaF_2	ββ and DM	concluded	LNGS
ELEGANT VI	CaF_2	ββ and DM	concluded	Oto

Alkali halide scintillators are also widely used for nuclear medicine (NaI(Tl)), high-energy physics (CsI(Tl), CsI pure), geophysics (NaI(Tl), CsI(Na)), environment and security control (CsI(Tl), ^{6}LiI(Eu)) and others. In recent years new Li-based halide scintillators (for example, $LiBaF_3$) are under investigation.

Leader role in investigation of rare processes

Scintillators by DAMA and by DAMA+INR-Kiev mainly to investigate $\beta\beta$ decay modes with source=detector approach

CaF_2(Eu)	Bicron/Crismatec(Saint Gobain)	NPA 705 (2002) 29; NPB 563 (1999) 97; Astrop. Phys. 7 (1997) 73
CeF_3	Crystal Clear coll. or China	NIMA 498 (2003) 352; NCIMA 110 (1997) 189
BaF_2	China or Bicron/Saint Gobain	NIMA525 (2004) 535
LiF	ISMA – Ukraine	NPA 806 (2008) 388; PLB 711 (2012) 41
$ZnWO_4$	Ukraine\Russia	JPG:NPP 38 (2011) 115107 NIM A 626-7 (2011) 31; NPA 826 (2009) 256 PLB 658 (2008) 193
($LaCl_3$(Ce)	Saint Gobain	Ukr. J. of Phys.51 (2006) 1037 NIMA555 (2005) 270)
$CeCl_3$	Iltis/Saint Gobain	NPA 824 (2009) 101; JPG:NPP 38 (2011) 015103
Li_2MoO_4	Russia	NIMA 607 (2009) 573
$Li_6Eu(BO_3)_3$	Ukraine	NIMA572 (2007) 734
$CdWO_4$	Ukraine development towards:	EPJA 36 (2008), 167; PRC 76 (2007) 064603
$^{106}CdWO_4$	Russia	NIM A 615 (2010) 301; PRC 85 (2012) 044610
$^{116}CdWO_4$	Russia	JINST 6 (2011) P08011
SrI_2(Eu)	Ukraine	NIM A 670 (2012) 10

and also polycrystalline powder.

ZnS(Ag)	Saint-Gobain	MPLA 27 (2012) 1250031

High radio-purity reachable in crystal scintillators?

- Identification of materials sources
- All involved materials selection within those potentially available at time of developments/production by:
    - Low background HPGe located deep underground
    - Mass and atomic spectrometry with high sensitivity
    - Neutron activation
- Devoted study of the presence of standard (U, Th, K) and non-standard contaminants
- Chemical/physical purification of the selected materials
- Selection of the more suitable growing process
- Additives selections
- Growing protocols
- Handling protocols
- Selection of the material other than crystal compounds
- Protocols for the assembling, the transport, the storage, the installation and maintenance in running conditions
- Prototypes tests deep underground

Necessary: many years,
many specific experience in the
specific detector, long dedicated time.
This kinds of development and measurements
themselves difficult experiments.

NO

OK

Produce detectors for Physics, but each one will have its own radio-purity + production differences….

Strength of crystal scintillators in low background applications

- Well known technology
- High duty cycle
- Large mass possible
- “Ecological clean” set-up; no safety problems
- Cheaper than other considered technique
- Relatively small underground space needed
- High radiopurity by selections, chem./phys. purifications, protocols reachable
- Well controlled operational condition feasible
- Neither re-purification procedures nor cooling down/warming up (reproducibility, stability, ...)
- Possibility of high light response in many cases
- Effective routine calibrations feasible in the same conditions as production runs
- Absence of microphonic noise
- Possibility of application both in passive and active source approaches as well as with coincidence/anticoincidence techniques
- Use of enriched materials
- Many isotopes and decay modes explorable

High benefits/cost

An example in case of sensitivity on ββ processes

Maximize abundance and detection efficiency

Enrichment, improving detector features

Maximize Target Mass and exposure Time

Multi-detectors set-up, scale up

$$T_{1/2}^{0\nu} \propto a\varepsilon\sqrt{\frac{Mt}{\Delta E B}}$$

Maximize Energy Resolution

Improving detector features

Minimize Background:

ULB techniques, segmented set-ups

Examples of isotopes which can be investigated by crystal scintillators with source=detector approach for ββ processes

Isotope	Nat. Ab. (%)	Q (keV)	Decay Mode	Scintillator
^{64}Zn	48.63	1095.7	$EC\beta^+$,2EC	$ZnWO_4$, $CdWO_4$
^{70}Zn	0.62	998.5	$2\beta^+$	$ZnWO_4$, $CdWO_4$
^{180}W	0.12	144	2EC	$ZnWO_4$, $CdWO_4$, $PbWO_4$
^{186}W	28.43	489.9	$2\beta^-$	$ZnWO_4$, $CdWO_4$, $PbWO_4$
^{106}Cd	1.25	2771	$2\beta^+$, $EC\beta^+$	$^{106}CdWO_4$
^{108}Cd	0.89	269	2EC	$CdWO_4$
^{114}Cd	28.73	536.8	$2\beta^-$	$CdWO_4$
^{116}Cd	7.49	2805	$2\beta^-$	$^{116}CdWO_4$
^{40}Ca	96.941	193.78	2EC	CaF_2, $CaMoO_4$
^{46}Ca	0.004	990.4	$2\beta^-$	CaF_2, $CaMoO_4$
^{48}Ca	0.187	4272	$2\beta^-$	CaF_2, $CaMoO_4$
^{136}Ce	0.185	2419	$2\beta^+$,$EC\beta^+$	$CeCl_3$, CeF_3, $CeBr_3$
^{138}Ce	0.251	693	2EC	$CeCl_3$, CeF_3, $CeBr_3$
^{142}Ce	11.114	1416.9	$2\beta^-$	$CeCl_3$, CeF_3, $CeBr_3$
^{130}Ba	0.106	2611	$2\beta^+$,$EC\beta^+$, 2EC	BaF_2, $BaCl_2$(Eu), BaI_2(Eu)
^{92}Mo	14.84	1649	$EC\beta^+$,2EC	$PbMoO_4$, $LiMoO_4$, $CaMoO_4$
^{100}Mo	9.63	3034	$2\beta^-$	$PbMoO_4$, $LiMoO_4$, $CaMoO_4$
^{84}Sr	0.56	1786.8	$EC\beta^+$	$SrCl_2$, SrI_2(Eu)

Radio-purity of crystal scintillators

arXiv:0903.1539[nucl-ex]

Radioactive contamination of crystal scintillators (mBq/kg)

Scintillator	Total α activity (U + Th)	^{228}Th	^{226}Ra	^{40}K	Particula radioactivity
$CaWO_4$	400	0.6	5.6	≤ 12	
	930 [a]	< 0.2	7		
$ZnWO_4$	0.2	0.002	0.002	≤ 0.4	0.5 (^{65}Zn)
$CdWO_4$	0.3 – 2	< 0.003 – 0.039	< 0.004	0.3 – 3.6	558 (^{113}Cd)
$PbWO_4$	(53 – 79)×10^3	≤ 13	≤ 10		(53 – 79)×10^3 (^{210}Pb)
$PbWO_4$ (from ancient lead)					≤ 4 (^{210}Pb)
$PbMoO_4$					(67-192)×10^3
$CaMoO_4$	≤ 10	0.04	0.13	≤ 3	
YAG:Nd	≤ 20				
BGO		< 0.4	< 1.2		7 – 3×10^3 (^{207}Bi)
GSO(Ce)	40	2.3	0.3	≤ 14	1200 (^{152}Gd)
		100	1.3		
NaI(Tl)		0.014	0.045		
	1.7	0.02	0.2		
	0.08	0.009	0.012	< 0.6	
CsI(Tl)		$\boxed{0.002}$	$\boxed{0.008}$	< 0.3	14 ^{134}Cs) 6 (^{137}Cs)
CaF_2(Eu)	8	0.13	1.3	≤ 7	10 (^{152}Eu)
		0.1	1.1		
CeF_3	3400	1100	≤ 60	≤ 330	
BaF_2		400	1400		
$LaCl_3$(Ce)		≤ 0.4	≤ 34		21×10^3 (^{138}La)
LuI_3					1.7×10^7 (^{176}Lu) [b]

[a] Estimated from the spectra presented in Fig. 13 of Ref. [37].
[b] Calculated value based on the half-life of ^{176}Lu: $T_{1/2}$ = 3.78×10^{10} y, its isotopic abundance (2.59%) [25], and chemical formula of the LuI_3 compound.

DAMA+INR Kiev

INR Kiev &
DAMA+INR Kiev

DAMA/LIBRA

KIMS

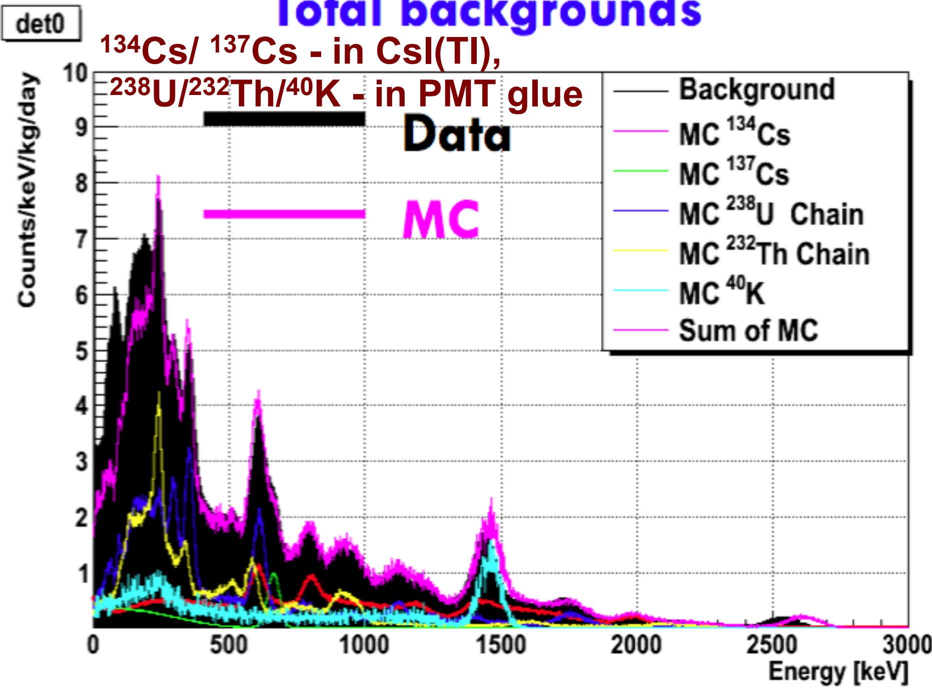
DAMA

ELEGANT VI

The highest sensitivity to measure internal contamination of crystal scintillators can be achieved in low background measurements where a scintillator is operating as a detector

- Time-amplitude analysis
- Pulse-shape discrimination
- Energy spectra analysis

Total backgrounds

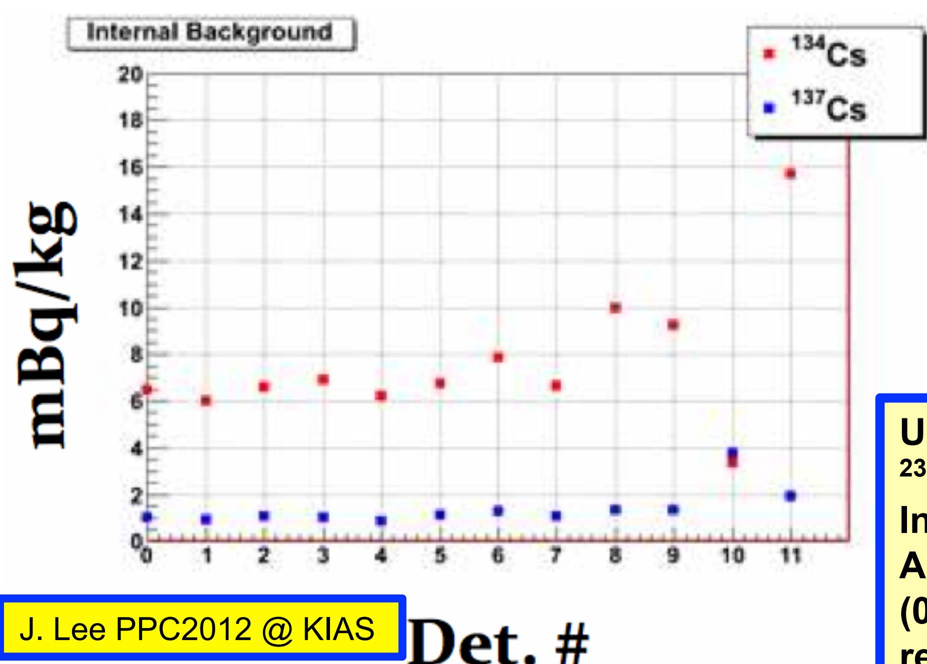


KIMS exp: 12 CsI(Tl) crystals, 8.7 kg, 8x8x30 cm^3=103.4 kg, in the Yangyang underground lab (700m depth) in Korea.

Internal background by alpha events

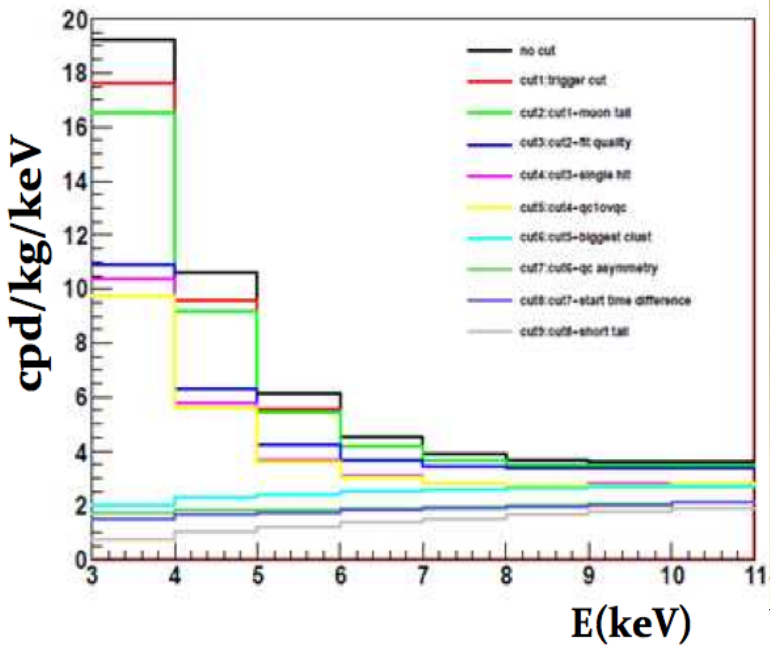
	Activity of Nuclide (μBq/kg)						
detector	^{238}U	^{234}U^a	^{222}Rn^b	^{210}Po	^{232}Th	^{228}Th^c	^{220}Rn^d
0	109.05	246.36	7.89	75.37	23.51	25.61	7.89
1	84.79	220.59	11.91	16.82	34.77	33.21	11.73
2	101.96	226.11	7.58	27.11	29.63	36.37	9.61
3	15.32	45.29	5.34	23.21	7.11	17.72	6.15
4	33.19	97.12	4.18	2.35	17.23	9.43	4.71
5	14.99	44.68	3.05	8.05	6.53	7.40	3.70
6	22.60	65.86	11.15	2.83	9.62	22.65	9.62
7	73.64	165.87	7.98	1.81	11.63	21.57	8.73
8	14.03	35.07	5.22	11.72	7.97	14.73	6.03
9	2.11	19.60	2.11	5.79	7.26	7.25	3.63
10	6.42	22.64	6.42	271.30	3.53	6.95	3.48
11	7.83	19.30	0.97	28.81	4.49	3.84	1.92

^{a} ^{234}U+^{230}Th+^{226}Ra. ^{b} ^{222}Rn and its alpha decay progenies.
^{c} ^{228}Th+^{224}Ra. ^{d} ^{220}Rn and its alpha decay progenies.



J. Lee PPC2012 @ KIAS

U chain probably broken, spread in contamination of ^{238}U,^{234}U.
In NIMA571(2007)644 and in Y.Kim "Closing in on DM", Aspen2013 ^{238}U and ^{232}Th concentrations are quoted (0.75 ± 0.23) and (0.38 ±0.07) ppt (~9 and ~2 μBq/kg respectively)

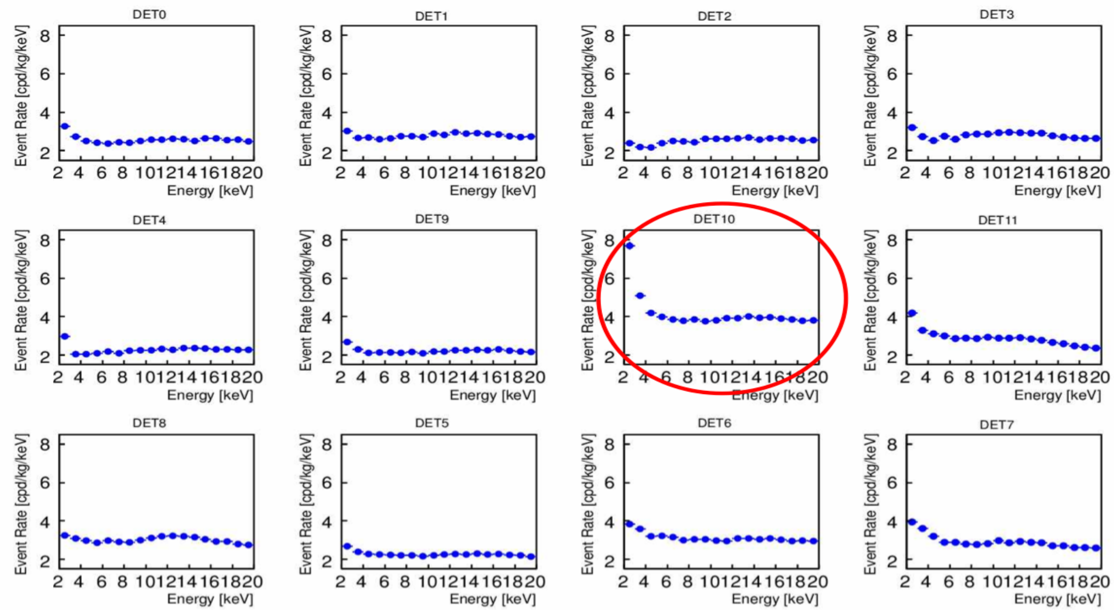


2012-11-05 [Single hit events]

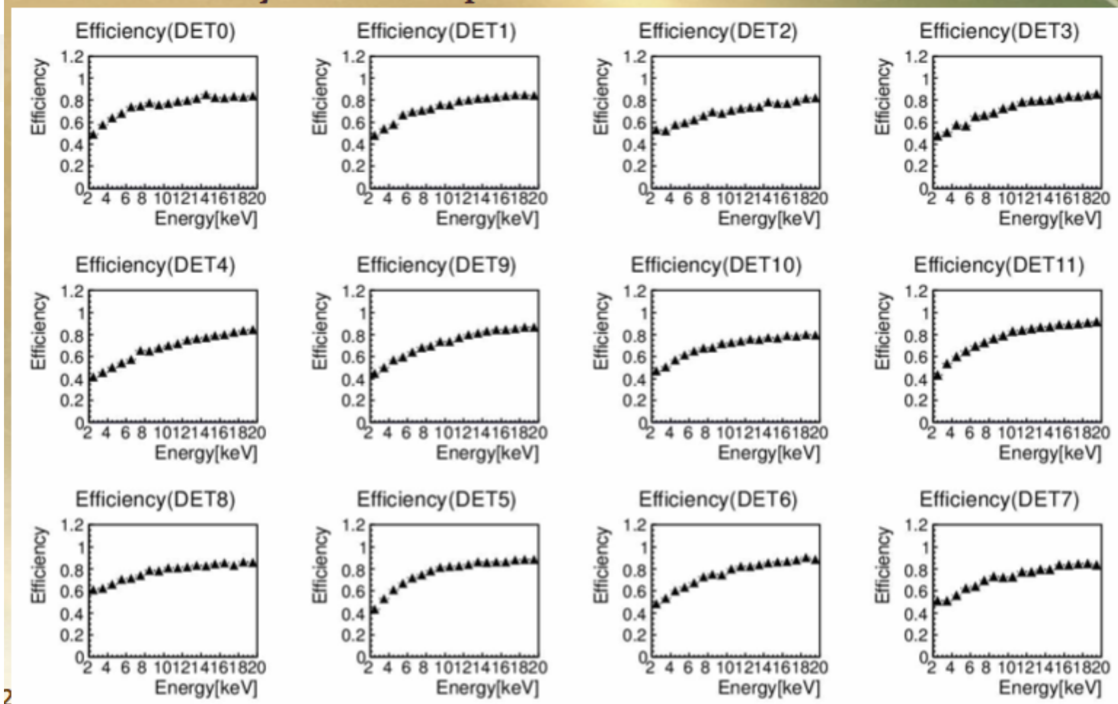
DAQ rate : < 6 Hz
High raw rate at low energy
Efficiency < 1 still @20 keV
Strong cuts still @ 11 keV

KIMS:J. Lee PPC2012 @ KIAS

- Single hit events after all cuts and efficiency correction (except Det10 for its high background rate)



- Cut efficiency from multiple hit data



An example in DAMA experience: which NaI(Tl)?

Qualification of NaI(Tl)	^{nat}K (ppb)	U (ppt)	Th (ppt)	Method of production
Standard	2000	< 500	< 500	Bridgman standard growth
Low Background	< 500	< 500	< 500	K Selected batches Bridgman growth
Very Low Background	< 100	< 50	< 50	K, U+Th Selected batches + Kyropoulos growth
Ultra Low Background (project Gran Sasso)	<< 40	< 5	< 5	Purified raw materials N and TlI + Crafted Kyropoulos growth + Handling protocol

"There are two necessary conditions imposing the choice of such inorganic scintillator for ultra-low background:

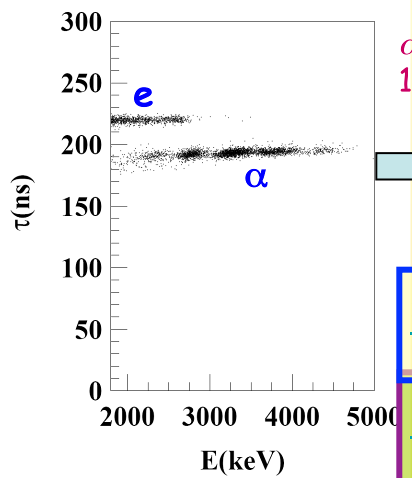
a) the matrix elements of composition do not contain long lived isotopes
b) the level of impurities generating natural radioactivity incorporating the crystals have to be sufficiently low (better than ppt)"

Kyropoulos crystallization process (in platinum crucible when growing for final detectors) acts as an additional considerable purification step

determination with the highest sensitivity by measurements with the detectors deep underground

VLB and ULB: long and delicate work, far from standard commercial production

Some on residual contaminants in DAMA NaI(Tl) detectors



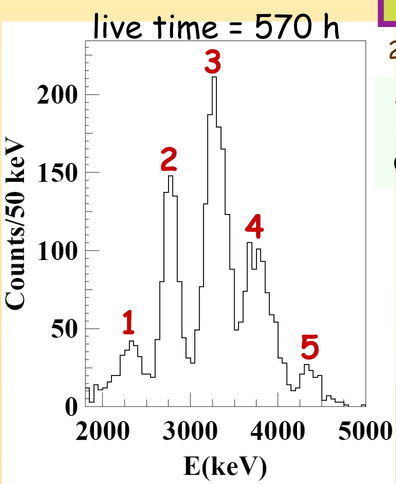
α/e pulse shape discrimination has practically 100% effectiveness in the MeV range

The measured α yield in the new DAMA/LIBRA detectors ranges from 7 to some tens α/kg/day

Second generation R&D for new DAMA/LIBRA crystals: new selected powders, physical/chemical radiopurification, new selection of overall materials, new protocol for growing and handling

^{232}Th residual contamination From time-amplitude method. If ^{232}Th chain at equilibrium: it ranges from 0.5 ppt to 7.5 ppt

^{238}U residual contamination First estimate: considering the measured α and ^{232}Th activity, if ^{238}U chain at equilibrium $\Rightarrow$ ^{238}U contents in new detectors typically range from 0.7 to 10 ppt

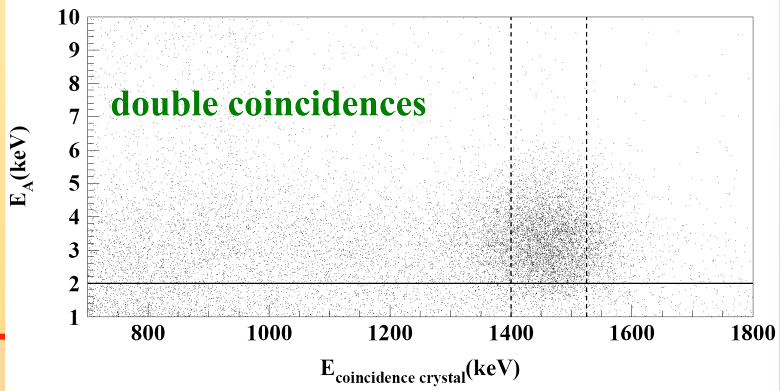


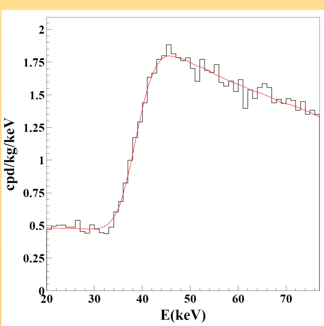
^{238}U chain splitted into 5 subchains: $^{238}U \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{210}Pb \rightarrow {}^{206}Pb$

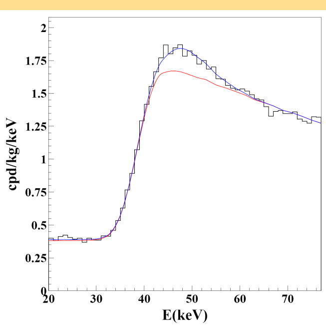
Thus, in this case: (2.1±0.1) ppt of ^{232}Th; (0.35 ±0.06) ppt for ^{238}U
and: (15.8±1.6) μBq/kg for ^{234}U + ^{230}Th; (21.7±1.1) μBq/kg for ^{226}Ra; (24.2±1.6) μBq/kg for ^{210}Pb.

^{nat}K residual contamination

The analysis has given for the ^{nat}K content in the crystals values not exceeding about 20 ppb







^{129}I and ^{210}Pb

$^{129}I/^{nat}I \approx 1.7\times10^{-13}$ for all the new detectors

^{210}Pb in the new detectors: (5 − 30) μBq/kg.

No sizeable surface pollution by Radon daugthers, thanks to the new handling protocols

For details and other information see NIMA592(2008)297

Light collection, Energy Threshold, Energy resolution

	Threshold (keV)	Rate @ thr (cpd/kg/keV)	σ/E	p.e./keV
ANAIS-0 NaI(Tl)	2-4	~10	10%60 keV	4.1-4.9
ANAIS-25(no LG)	-	-	5.7%@60 keV	13-16
NAIAD NaI(Tl)	2	10	8.5%@60 keV	4.6-9
DM-ICE-17	~7	6	-	4.7-6.1
PICO-LON	-	-	10.2@60 keV	-
DAMA/LIBRA NaI(Tl)	2	1	7.5%@60 keV	5.5-7.5
DAMA/LIBRA phase II	<2	-	6.7%@60 keV	6-10
LSM NaI(Tl)	2	10	5%@60 keV	9
KIMS Cs(Tl)	3	3	8.2%@60 keV	5
CaF_2(Eu) Kamioka	2	10	34%@5.9 keV	4
DAMA-CaF_2(Eu)	4	8	11%@60 keV	-

The light collection optimization is user dependent and has to be made on a case by case basis. A good light collection scheme should:

- maximize the number of photons extracted
- keep a good linearity and uniformity of the response.

some impurities and imperfections present in a crystal at the level of a few ppm may influence the optical quality and increase the afterglow.

Even more for DM application

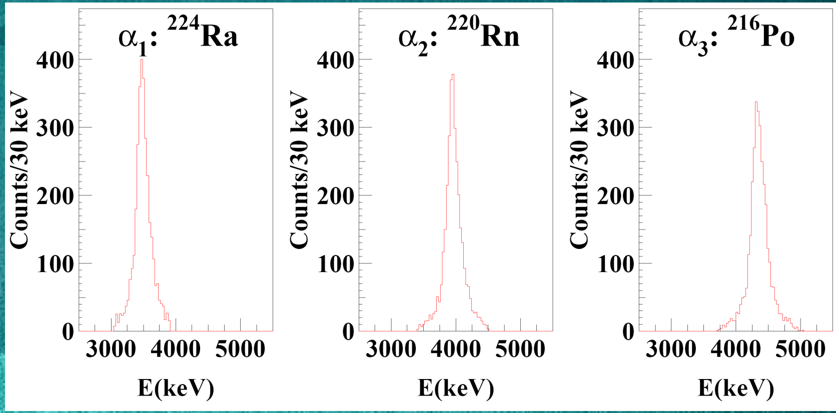
In DM investigation:
high number of phe/keV is important to reach low threshold
uniformity and linearity are fundamental to have the full control of the detector response

An example: DAMA/LIBRA

Uniformity of the light collection

- Absence of dead spaces in the light collection: no significant variations of the peak position and energy resolution when irradiating the whole detector with high-energy γ sources (e.g., ^{137}Cs) from different positions.

α peaks at high energy and their energy resolutions are well compatible with those expected for γ calibration (ex: energy resolution σ = (75±3) keV for α_1, for γ's is 72 keV).

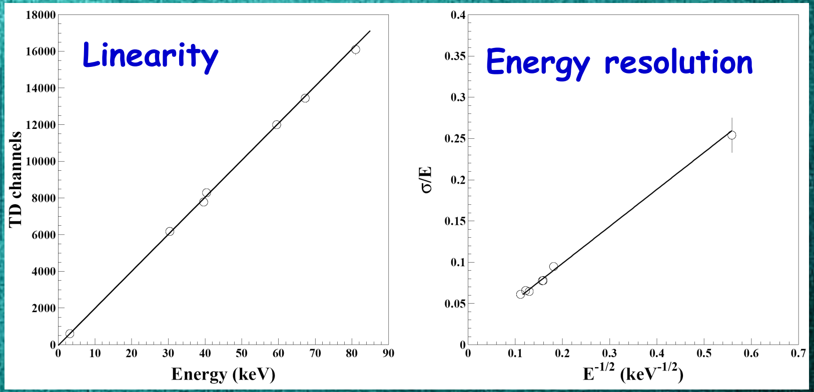


All this supports the uniformity of the light collection within 0.5%.

Linearity of the light collection

Low energy: various external γ sources (^{241}Am, ^{133}Ba) and internal X-rays or γ's (^{40}K, ^{125}I, ^{129}I), routine calibrations with ^{241}Am

$$\frac{\sigma_{LE}}{E} = \frac{(0.448 \pm 0.035)}{\sqrt{E(keV)}} + (9.1 \pm 5.1) \cdot 10^{-3}$$

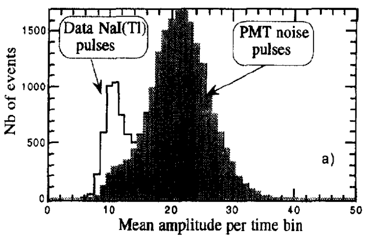


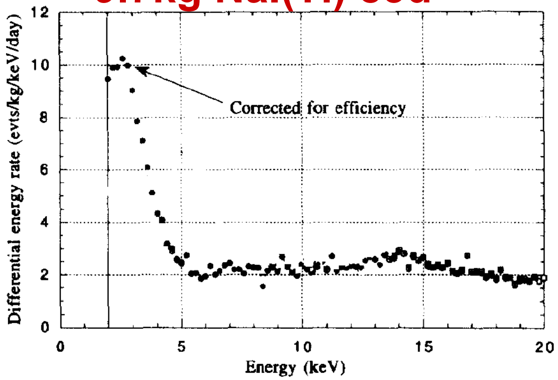
Example of scintillation energy spectra measured by various NaI(Tl) detectors

- Shapes/scintillation rates quite different
- All of them cannot be easily/uniquely simulated by a MC code

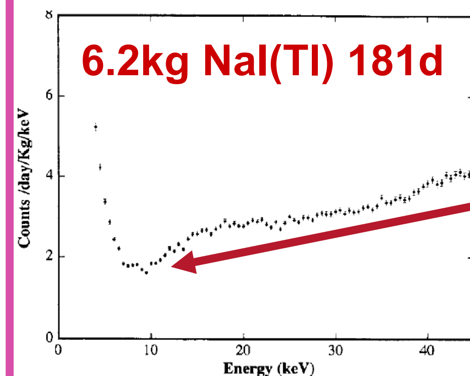
Modane 1999 Astrop.Phys.11(1999)287

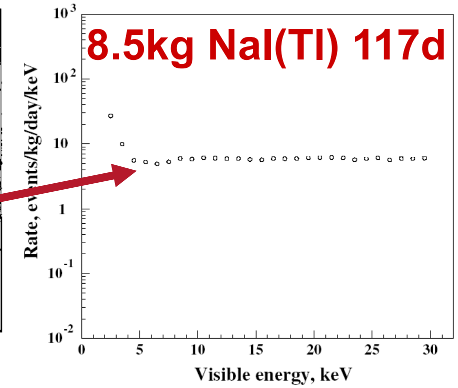
poor noise rejection although high n. of ph.e.
$\sigma/E=1.2\%/\sqrt{E(MeV)}$





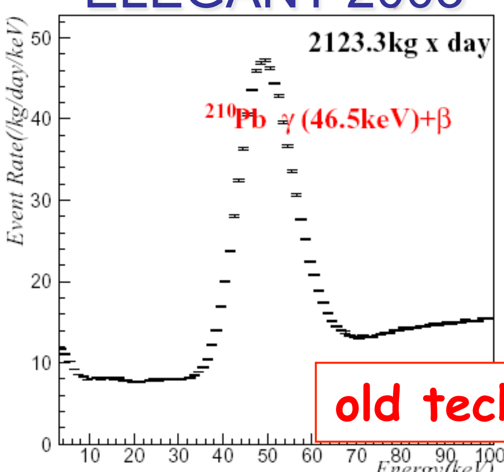
NAIAD 1996 NAIAD 2003





Astrop.Phys.5(1996)249 Astrop.Phys.19(2003)691

ELEGANT 2003

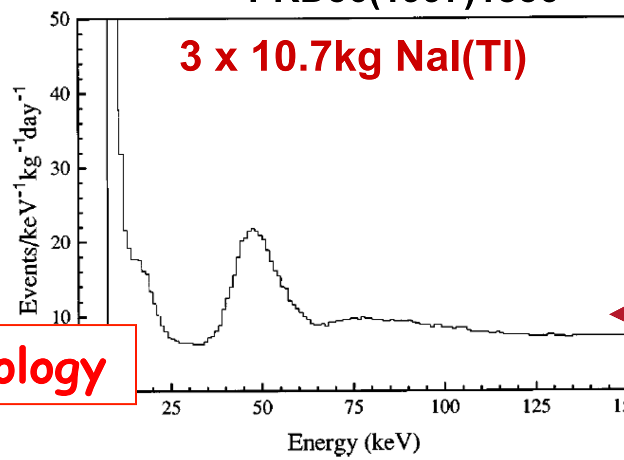


NDM2003

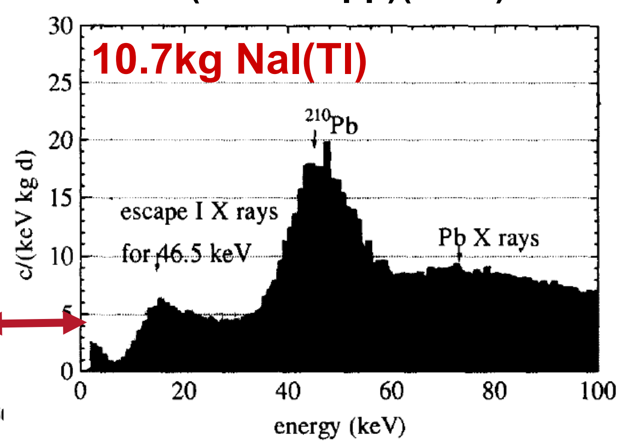
old technology

ANAIS

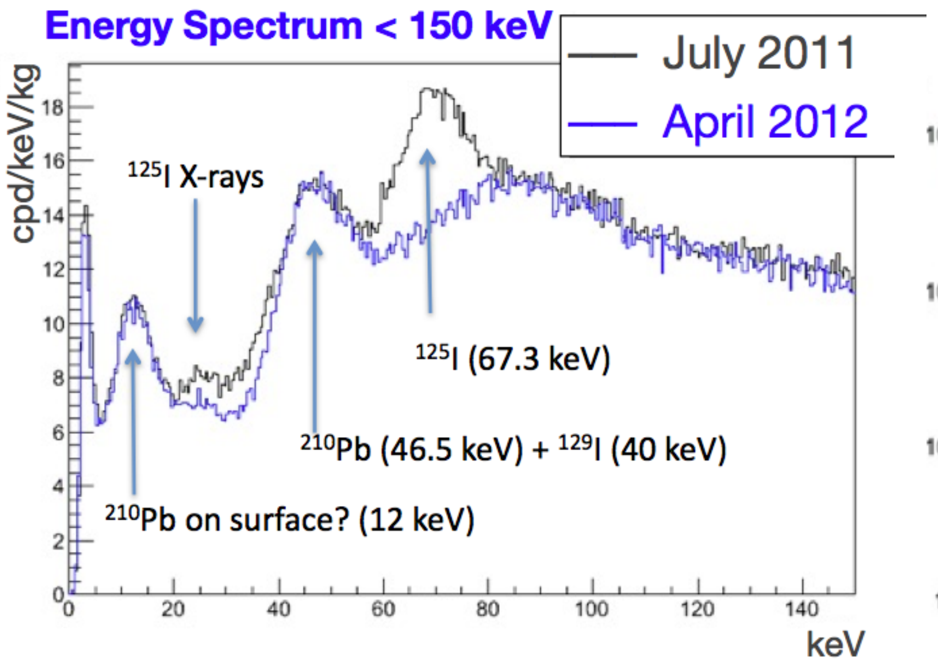
PRD56(1997)1856

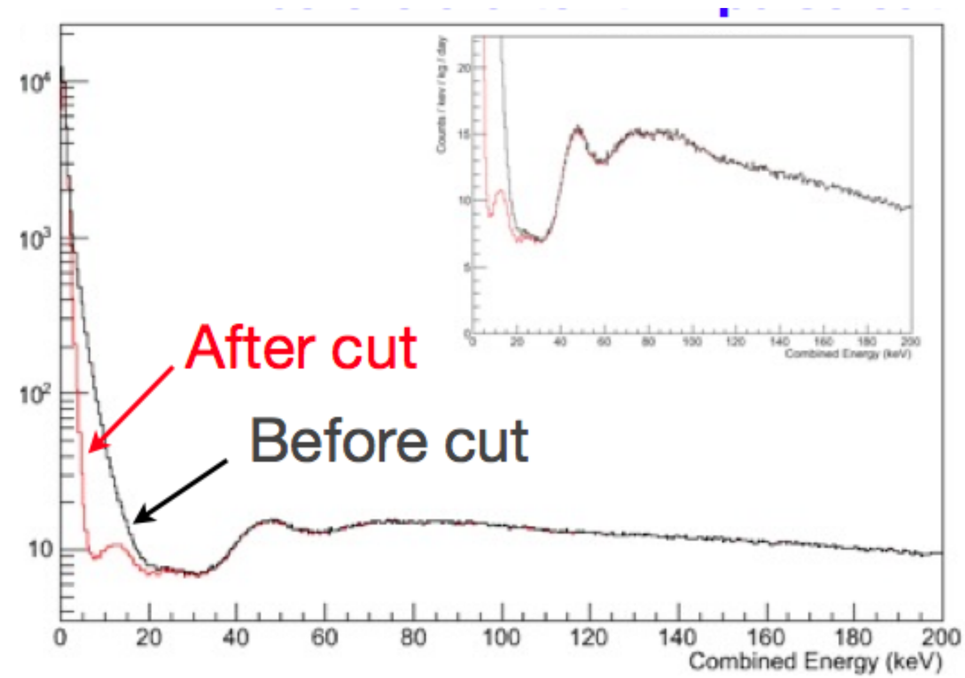


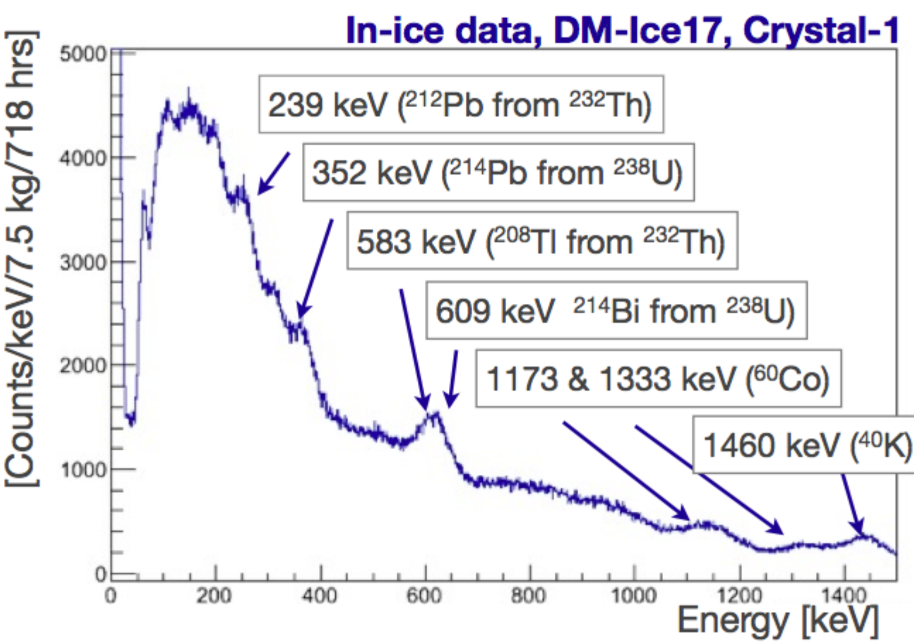
NPB114(Proc.Supp)(2003)111



DM-ICE-17 R.Maruyama, Aspen, Feb. 2013

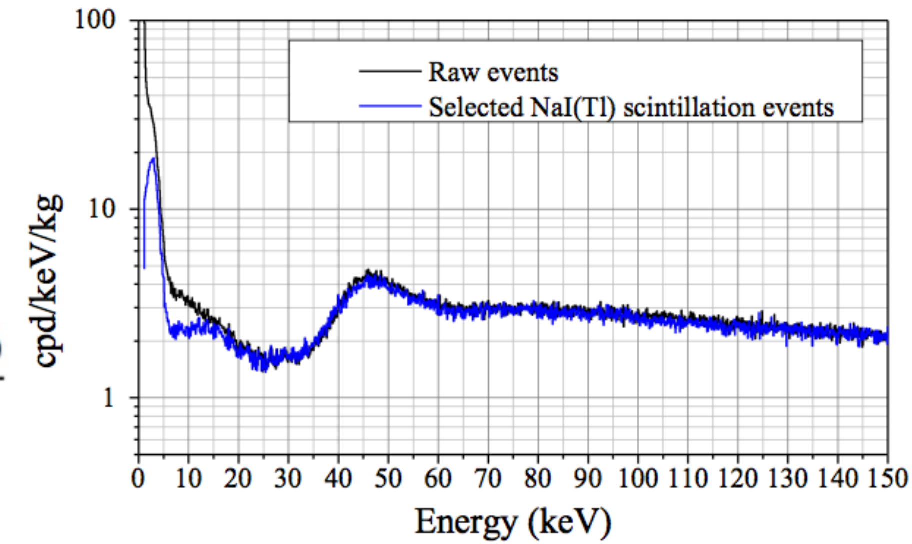






	DM-Ice-17 (μBq/kg)
U238	No info
U234	93
Th230	93
Ra226-Pb210	933
Bi210-Pb206	1680
Th232	214

Two NAIAD NaI Crystals (5”x5”, 8.5 kg)=17 kg
Operation since Feb. 2011
Data run from June 2011
Plans for new crystals up to 250 kg set-up



Astrop. Phys. 37 (2012) 60

identified radioactive isotopes bulk-distributed in the NaI crystal and considered in the simulation of the ANAIS-0 module.

Isotope	NaI crystal bulk activity (mBq/kg)
^{40}K	12.7 ± 0.5
^{129}I	0.96 ± 0.06
$^{238}U/^{234}U$	0.075 ± 0.005
^{230}Th	0.023 ± 0.007
^{226}Ra	0.098 ± 0.004
^{210}Pb	0.188 ± 0.005
^{232}Th	0.013 ± 0.005
^{228}Th	0.035 ± 0.003

ANAIS-0: 9.6 kg St. Gobain 25.4 x 10.2 x 10.2 cm^3 @Canfranc

250 kg **NEXT STEP ANAIS-25:**

SELECTED POWDER (AS):

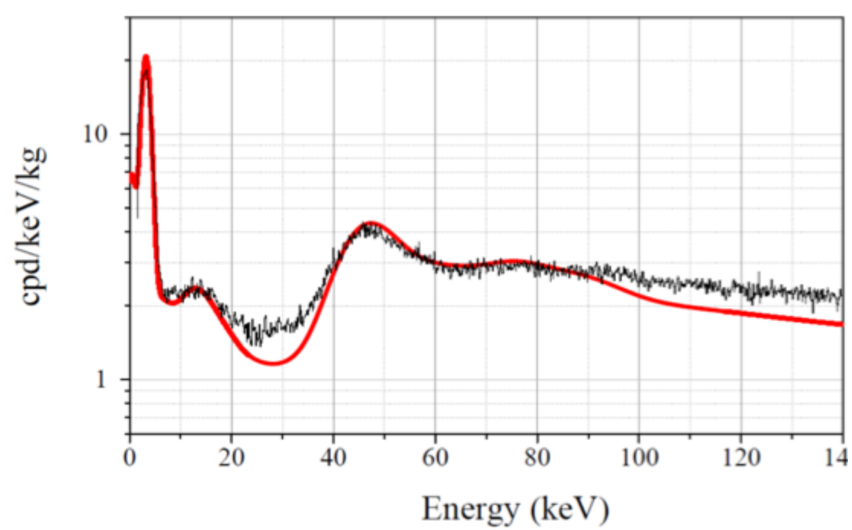
K (ppb)	U (ppb)	Th (ppb)
< 90	< 0.055	< 0.13

UPPER LIMITS AT 95% C.L.

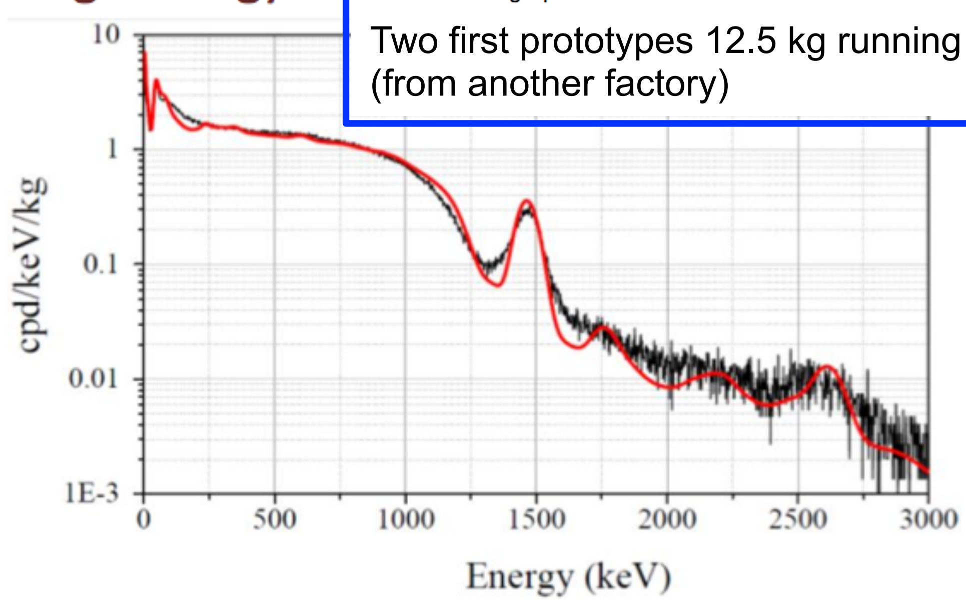
assuming equilibrium in U and Th chains

Two first prototypes 12.5 kg running (from another factory)

Low energy:



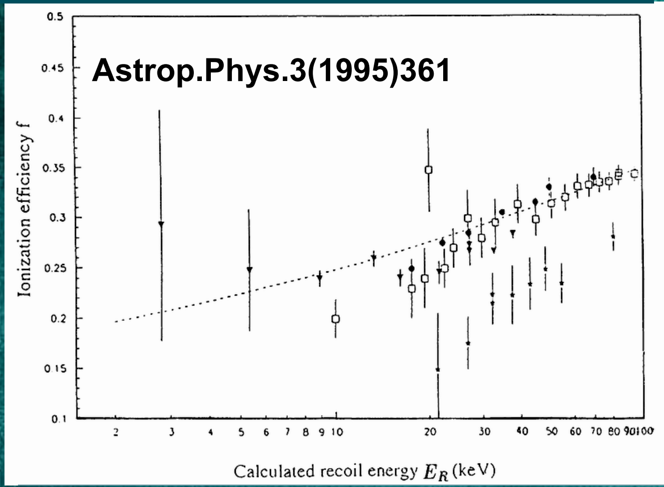
High energy:



M. Martínez, seminar@LNGS, Feb 2013

QUENCHING FACTOR

Ex. of different *q* determinations for Ge



differences are often present in different experimental determinations of q for the same nuclei in the same kind of detector

e.g. in doped scintillators q depends on dopant, on the impurities/trace contaminants, on specific light response, on growth procedures; in LXe e.g. on trae impucrities, on initial UHV, on presence of degassing/releasing materials in the Xe, on thermodynamical conditions, on possibly applied electric field, etc.

Some time increases at low energy in scintillators (dL/dx)

… and more

examples of q measurement in some detectors with neutrons

Nucleus/Detector	Recoil energy (keV)	q
NaI(Tl)	(6.5–97)	(0.30 ± 0.01) for Na
	(22–330)	(0.09 ± 0.01) for I
	(20–80)	(0.25 ± 0.03) for Na
	(40–100)	(0.08 ± 0.02) for I
	(4–252)	(0.275 ± 0.018) for Na
	(10–71)	(0.086 ± 0.007) for I
	(5–100)	(0.4 ± 0.2) for Na
	(40–300)	(0.05 ± 0.02) for I
CaF_2(Eu)	(30–100)	(0.06–0.11) for Ca
	(10–100)	(0.08–0.17) for F
	(90–130)	(0.049 ± 0.005) for Ca
	(75–270)	(0.069 ± 0.005) for F
	(53–192)	(0.11–0.20) for F
	(25–91)	(0.09–0.23) for Ca
CsI(Tl)	(25–150)	(0.15–0.07)
	(10–65)	(0.17–0.12)
	(10–65)	(0.22–0.12)
CsI(Na)	(10–40)	(0.10–0.07)
Ge	(3–18)	(0.29–0.23)
	(21–50)	(0.14–0.24)
	(10–80)	(0.18–0.34)
	(20–70)	(0.24–0.33)
Si	(5–22)	(0.23–0.42)
	22	(0.32 ± 0.10)
Liquid Xe	(30–70)	(0.46 ± 0.10)
+PRC(2010)025808 &refs	(40–70)	(0.18 ± 0.03)
	(40–70)	(0.22 ± 0.01)
Bolometers	(80-130)	0.87±0.10
	(20-100)	0.91±0.03±0.04

Quenching factor is a relevant experimental parameter for DM candidates inducing nuclear recoils. It has to be considered in all kinds of detectors. In additional the channelling effect has to be included when dealing with q in crystals (Eur. Phys. J. C 53(2008)205,*J. Phys.: Conf. Ser.* 203(2010)012042)). See also expectations by phenomenological arguments in Astrop.Phys.33(2010)40.

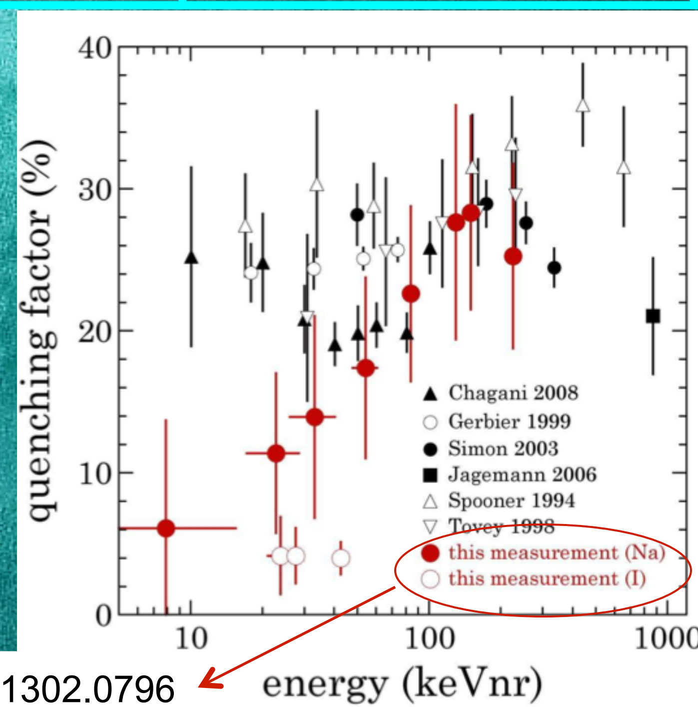
direct measurements of q. f. are performed with reference detectors that in some cases have features quite different from the detectors used in the running conditions.
The nature of these measurements, the parameterisation, the used neutron beam/ sources may not point out all the possible contributions or may cause uncertainties. Channeling could also play a role.

Example in case of DAMA	Na	I	Recoil energy range (keVee)	
Same method	0.30(1)	0.09(1)	6.5–97 (Na) 22–330 (I)	Phys. Lett. B389(1996)757
Same method	0.4 ± 0.2	0.05 ± 0.02	5–100 (Na) 40–300 (I)	Phys. Rev. C47(1993)R425
semi-empirical formula	from 0.65 to 0.55	from 0.35 to 0.17	2–100	Astrop. Phys. 33(2010)40

But: 1) the q.f. values depends on the specific crystal and detector; 2) no alpha light yield has been given for the used NaI(Tl) crystal for comparison; 3) etc…

In disagreement with other measured values, problem of crystal light response of inefficiency?

It is not correct to apply these results to all the NaI(Tl) detectors.



arXiv:1302.0796

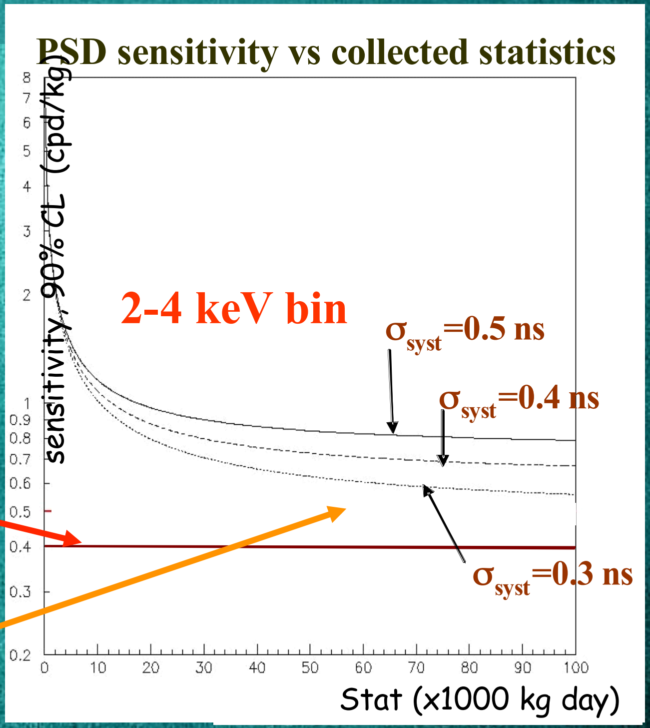
PSD

- **Any e.m. rejection technique is not a DM signature and it is blind to some candidates.**
- **Only a statistical discrimination is possible because, e.g., of tail effects from the two populations and of known concurrent processes (e.g., end-range alphas and neutrons induce signals indistinguishable from recoils) whose contribution cannot be estimated and subtracted in any reliable manner at the needed level of precision.**
- **PSD cannot safely be applied in the investigation of the DM annual modulation signature; in fact, the effect searched for (at level of few %) would be largely affected by the uncertainties associated to rejection procedure. On the other hand the annual modulation signature acts itself as an effective background rejection.**

  - **σ_{syst} limits the sensitivity of PSD when going towards large statistics; it depends on several factors: temperature variation during data taking, instrumental effects, stability of the selecting windows, ...**

Observed annual modulation effect in terms of S_0 for a particular model framework

It corresponds to 0.1 ^{0}C variation



Whatever e.m. rejection technique has similar drawbacks

Some comments on PSD in KIMS PRL 108 (2012) 181301

KIMS exp: 12 CsI(Tl) crystals, 8.7 kg, 8x8x30 cm^3, in the Yangyang underground lab (700m depth) in Korea; data from Sept 2009 to Aug 2010, exposure about 2.5×10^4 kg x d

Not discussed control on systematics (temperature, stability etc.), in fact:

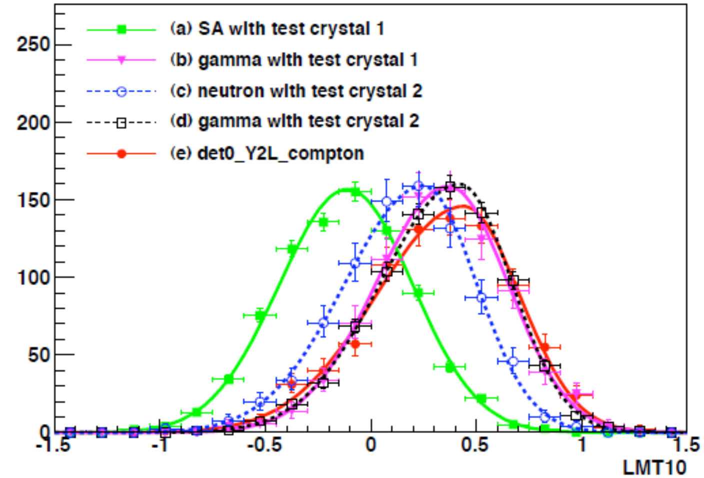
- Discrimination power of about 2-3 x 10^{-3} **claimed** (=10^{-2} cpd/kg/keV / 2-3 cpd/kg/keV (but raw rate strongly higher)); to have such discrimination power it is necessary the control of all the systematics at a much better level (**much below 10^{-3}**).

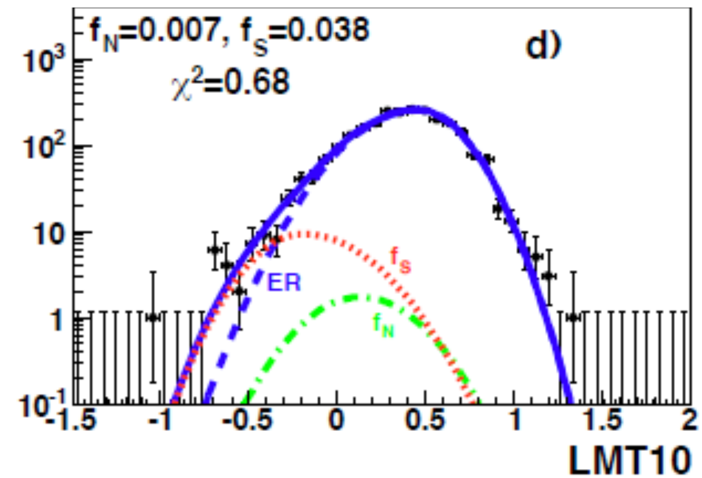
- Energy scale: no info about the validation of the 3 keV energy threshold (no calibration point close to the energy threshold, energy resolution not demonstrated at low energy).

- The stability of the efficiency and of the procedure to remove the PMT noise events not discussed

- The systematics effects for the SA (surface alphas) events not evaluated (their distribution among the detectors)

- It is written "the mean values of the ER logrmt10 distributions have an average value of 0.62 with a rms spread of 0.035 ...", but in Fig. 1 at different energy the mean value of one detector is about 0.4, that is more than 5 sigma away.





- It is clear from table in the paper and, in particular, from fig. 2d that the weights of the different contributions are about:
  (3-5)/ (0.05-0.10) /(0.01) for ER (electrons) / SA / recoils, but it is never mentioned if the whole PSD procedure can be reliably applied to disentangle the three contributions at so-low "extrapolated" energy.

The DAMA/LIBRA set-up ~250 kg NaI(Tl) (Large sodium Iodide Bulk for RAre processes)

As a result of a 2nd generation R&D for more radiopure NaI(Tl) by exploiting new chemical/ physical radiopurification techniques

DAMA/NaI (7 years) + DAMA/LIBRA (6 years) Total exposure: 425428 kg$\times$day = 1.17 ton$\times$yr data favor the presence of DM particles in the galactic halo at about 9σ C.L.

Residual contaminations in the new DAMA/LIBRA NaI(Tl) detectors: ^{232}Th, ^{238}U and ^{40}K at level of 10^{-12} g/g

JINST 7(2012)03009

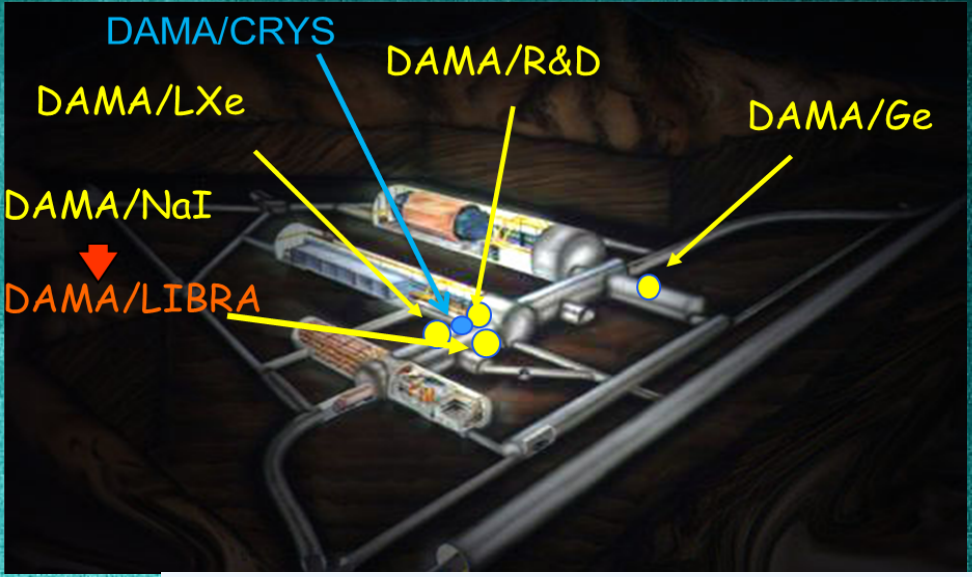
DAMA/LIBRA - phase 2

Continuously running with new PMTs with higher Q.E. with lower software energy threshold (below 2 keV).

Radiopurity, performances, procedures, etc.: NIMA592(2008)297, JINST 7 (2012) 03009
Results on DM particles, Ann. Modulation Signature: EPJC56(2008)333, EPJC67(2010)39
Related topics: PRD84(2011)055014, EPJC 72 (2012) 2064
Results on rare processes: PEP violation EPJC62(2009)327, CNC in I EPJC72(2012)1920

Search for rare processes with other set-ups

- **All set-ups running**
- **Wide program for many measurements with INR-Kiev (see also F. Danevich's talk on ^{106}Cd and ^{116}Cd)**



Some of the past results:

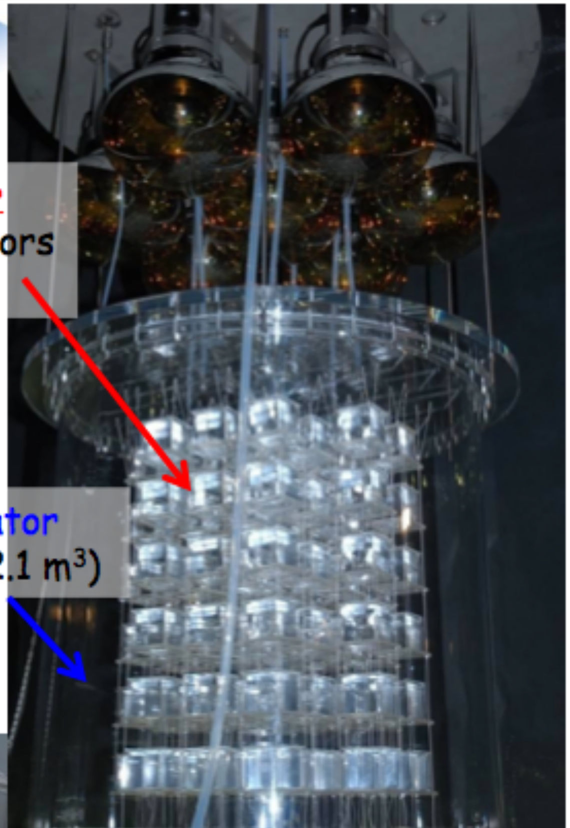
- 2β decay in ^{136}Ce, ^{138}Ce, ^{142}Ce, ^{40}Ca, ^{46}Ca, ^{48}Ca, ^{106}Cd, ^{108}Cd, ^{114}Cd, ^{116}Cd, ^{130}Ba, ^{64}Zn, ^{70}Zn, ^{180}W, ^{186}W with various low background scintillators (NPA705(2002)29, NIMA498(2003)352, NIMA525(2004)535, PLB658(2008)193, NPA826(2009)256, JPG:NPP38(2011)115107, EPJA36(2008)167, JPG: NPP38(2011)015103, PRC85(2012)044610, JINST6(2011)P08011)

- α decay in ^{142}Ce, in ^{nat}Eu (NPA789(2007)15), β decay in ^{48}Ca, in ^{113}Cd (PRC76(2007)064603)
- Cluster decay in $LaCl_3(Ce)$ (NIMA555(2005)270)
- CNC decay $^{139}La \rightarrow {}^{139}Ce$ (UJP51(2006)1037)

- RDs on low background scint. and PMTs
- 2β decay in ^{100}Mo (NPA846(2010)143), ^{96}Ru and ^{104}Ru (EPJA42(2009)171), ^{136}Ce and ^{138}Ce (NPA824(2009)101), ^{190}Pt and ^{198}Pt (EPJA47(2011)91), ^{156}Dy and ^{158}Dy (NPA859(2011)126), ^{184}Os and ^{192}Os (EPJA49(2013)24)
- Search for ^{7}Li solar axions (NPA806(2008)388, PLB711(2012)41)
- First observation of α decay of ^{190}Pt to the first excited level of ^{186}Os (PRC83(2011)034603)
- Qualification and meas. of many materials: e.g. $CdWO_4$, $ZnWO_4$(NIMA626-7(2011)31, NIMA615(2010)301), $Li_6Eu(BO_3)_3$ (NIMA572(2007)734), Li_2MoO_4 (NIMA607(2009)573), $SrI_2(Eu)$ (NIMA670(2012)10), $^{7}LiI(Eu)$ (NIMA704(2013)40)

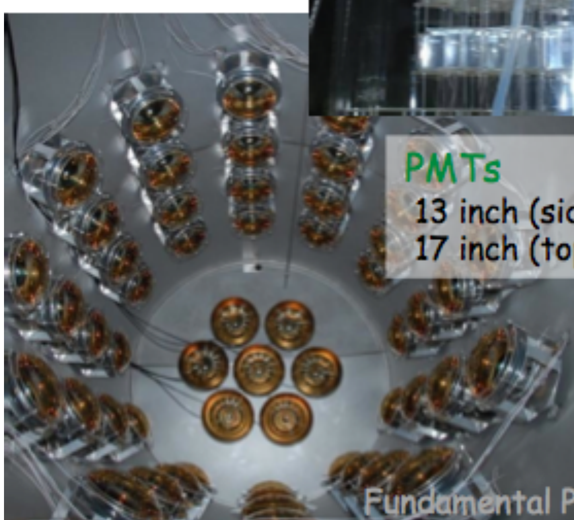
- ^{106}Cd, ^{116}Cd in progress (PRC85(2012)044610, JINST6(2011)P08011)
- ADAMO project: Study of the DM directionality approach with $ZnWO_4$ anisotropic detectors (EPJC73(2013)2276)

CANDLES III Detector



Main detector
CaF_2 scintillators (305kg)

Liquid scintillator
acrylic tank (2.1 m^3)



PMTs
13 inch (side) ; x 48本
17 inch (top & bottom) ; x 14本

- CaF_2 Module
  - CaF_2(Pure) ; 96 Crystal → 305 kg
  - WLS Phase ; 280 nm → 420 nm
    - Thickness ; 5 mm
    - Composition ; Mineral Oil+bis-MSB (0.1 g/L)
- Liquid Scintillator (LS)
  - 1.4 m ϕ x 1.4 m H
  - Volume ; 2.1 m^3 (1.65 ton)
  - Composition
    - Solvent ; Min
    - Solutes (WLS
- Acrylic Tank
  - Container for
- Water Buffer
  - Pure Water → Passive Shield
  (Pre,Final-filter, Chacoal-filter, UV-lamp, Ion-Exchanger
  - Distance PMT – LS ; 50 cm
- PMTs
  - 13 inch (Side) ; x 48本
  - 17 inch (Top and Bottom) ; x 14本

JoP:Conf.Ser.375(2012)042018
CaF_2 crystals: U-chain (Bi-214)
~36 μBq/kg (14±5 μBq/kg ;Best)
Th-chain(Rn-220) ~28 μBq/kg
(6±1 μBq/kg ;Best)

An interesting example for further developments: Ce isotopes

Searching for detectors to investigate ββ decay mode of Ce isotopes:

^{136}Ce (δ = 0.185%; Q = 2419keV): 2EC, ECβ^+, 2β^+
^{138}Ce (δ = 0.251%; Q = 693 keV): 2EC
^{142}Ce (δ = 11.114% ; Q = 1416.7keV): 2β^-

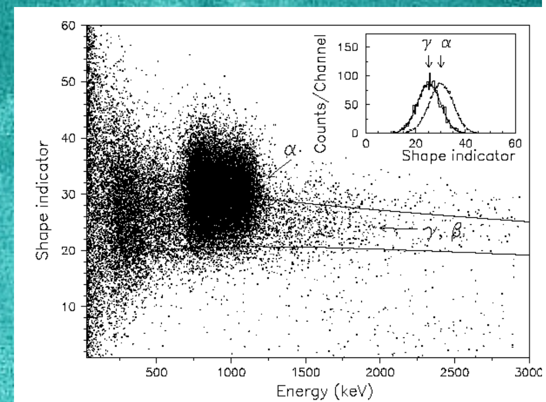
CeF_3 — NIMA 498 (2003) 352
$CeCl_3$ — NPA 824 (2009) 101; JPG:NPP 38 (2011) 015103

CeF_3 crystal scintillators

(2 x 2 x 2) cm³, mass 49.3 g, produced in China; E_{thr} = 20 keV; σ/E=22% at 122 keV

(14 x 2 x 2) cm³ , mass 345 g, produced by Preciosa-Crytur; E_{thr} = 150 keV; σ/E=29% at 662 keV

(2.2 x 2.2 x 2.5) cm³ , mass 74.5 g, produced in China; E_{thr} ~ 20 keV; σ/E=19% at 122 keV

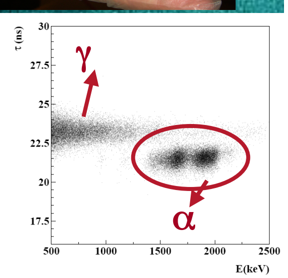


Radioactive contaminations in the CeF_3 crystal scintillator obtained by fitting the experimental spectrum by the background model. The derived in this way activity of ^{232}Th 55(30) mBq/kg is consistent with the value of 37(16) mBq/kg obtained by the pulse-shape analysis of the data

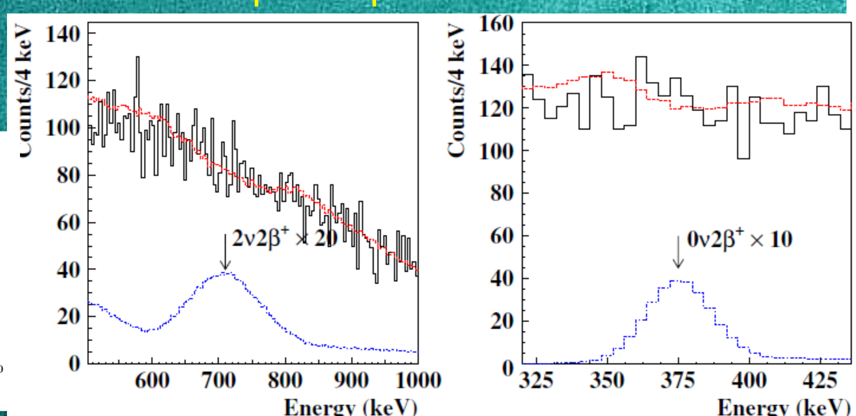
Chain	Source	Activity (mBq/kg)
^{232}Th	^{232}Th	55(30)
	^{228}Ra	890(270)
	^{228}Th	1010(10)
^{238}U	^{238}U	⩽70
	^{234}U	⩽60
	^{230}Th	⩽60
	^{226}Ra	⩽60
	^{210}Pb	⩽280
^{235}U	^{235}U	⩽40
	^{231}Pa	⩽50
	^{227}Ac	⩽20
	^{40}K	⩽330
	^{138}La	⩽60
	^{176}Lu	⩽20
	^{147}Sm	⩽80

$CeCl_3$ crystal scintillators

$CeCl_3$ (∅13×13 mm, 6.9 g)
Live time: 1638 h



2ν2β⁺ e 0ν2β⁺ in ^{136}Ce



Transition	Decay channel	Decay mode	Exp. $T_{1/2}$ limit (yr) Present work	Previous results
^{36}Ce → ^{136}Ba	$2\beta^+$	0ν	$> 0.7(2.5) \times 10^{17}$	$> (6.9) \times 10^{17}$ [9]
		2ν	$> 0.9(1.8) \times 10^{16}$	$> 1.8 \times 10^{16}$ [10]
	$\varepsilon\beta^+$	0ν	$> 0.9(2.5) \times 10^{17}$	$> 3.8 \times 10^{16}$ [10]
		2ν	$> 2.4(5.4) \times 10^{16}$	$> 2.6 \times 10^{15}$ [11]
	$2K$	0ν	$> 3.0(3.8) \times 10^{16}$	$> 6.0 \times 10^{15}$ [10]
		2ν	$> 3.2(4.2) \times 10^{16}$	$> 2.7 \times 10^{16}$ [8]
^{38}Ce → ^{138}Ba	$2K$	0ν	$> 3.6(4.7) \times 10^{16}$	$> 1.9 \times 10^{15}$ [11]
		2ν	$> 4.4(5.7) \times 10^{16}$	$> 3.7 \times 10^{16}$ [8]
^{42}Ce → ^{142}Nd	$2\beta^-$	0ν	$> 0.7(1.6) \times 10^{19}$	$> (1.5) \times 10^{19}$ [9]
		2ν	$> 1.4(3.0) \times 10^{18}$	$> 1.6 \times 10^{17}$ [10]

Example of the study of new kind of scintillator detector: $SrI_2(Eu)$ crystal scintillator:

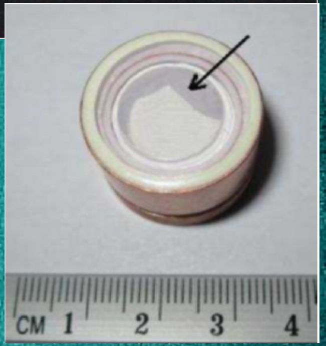
NIMA670(2012) 10

- High light output (>100000 γ/MeV)
- Good energy resolution (~3% at 662 keV)
- Absence of natural long-living radioactive isotopes.

Properties of $SrI_2(Eu)$ crystal scintillators.

Property	Value
Density (g/cm^3)	4.5–4.6
Melting point (°C)	515
Structural type	Orthorhombic
Index of refraction	1.85
Wavelength of emission maximum (nm)	429–436
Light yield (photons/MeV)	(68–120) $\times 10^3$
Energy resolution (FWHM, %) for 662 keV γ of ^{137}Cs	2.6–3.7
Scintillation decay time (μs) under X-ray/γ ray excitation at 300 K	0.6–2.4

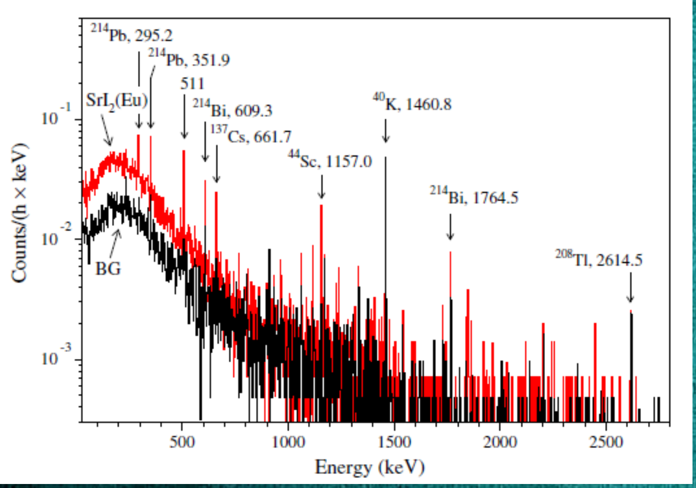
- A scintillator crystal doped by 1.2% in Eu and with a nearly cylindrical shape (13 x 11 mm; 6.6 g mass) produced by Stockbarger growth technique



- Preliminary measurement in the low background set-up installed at sea-level at INR, Kyiv, Ukraine:
  - detector performances
  - α/β discrimination

Measurement in the Ge facility at LNGS

Measured during 706 h with ULB-HPGe γ ray spectrometer to investigate background and put new limits on 2β decay of ^{84}Sr ($Q_{2\beta}$. 1787.4. keV)



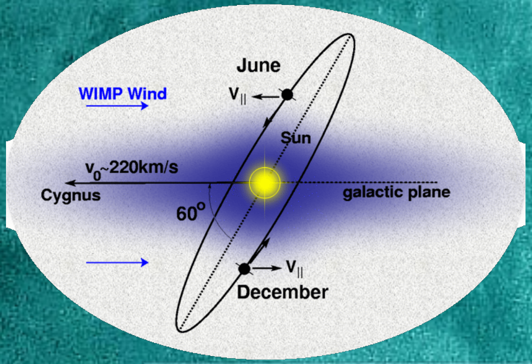
- Potentiality of $SrI_2(Eu)$ to the search for the ^{84}Sr 2β decay demonstrated for the first time (crystal mass= 6.6 g; $\delta_{^{84}Sr}$=0.56 (1)% ; measuring time = 101.52 h)
- New/improved half-life limits on 2ε and $\varepsilon\beta^+$ decay modes in ^{84}Sr at level $T_{1/2}$ ~ 10^{15}-10^{16} yr
- With higher mass crystal and longer meas. time high sensitivity expected

A R&D of $SrI_2(Eu)$ crystal scintillators is in progress

The ADAMO project: Study of the directionality approach with ZnWO4 anisotropic detectors

Eur. Phys. J. C 73 (2013) 2276

Directionality: study of the correlation between the Earth motion in the galactic rest frame and the arrival direction of those Dark Matter (DM) candidates inducing just nuclear recoils



- Nuclear recoils would be strongly correlated with the DM impinging direction
- This effect can be pointed out through the study of the variation in the response of **anisotropic scintillation detectors** during sidereal day

- **light output** and **pulse shape** of **$ZnWO_4$** depend on the direction of the impinging particles with respect to the crystal axes
- These **anisotropic features** provide two independent ways to exploit directionality, overcoming the difficulties of TPC detectors due to the very short track of recoils

Complementary information to those by DAMA/LIBRA for some aspects

ZWO (699g, ISMA)
Contamination (mBq/kg)

^{232}Th	< 0.1
^{228}Ra	< 0.05
^{228}Th	0.002(1)
^{227}Ac	<0.003
^{238}U+^{234}U	<0.08
^{230}Th	<0.07
^{226}Ra	0.002(1)
^{210}Po	<0.06
^{40}K	<0.4
^{65}Zn	0.5(1)
α activity	0.18(3)

Development of $ZnWO_4$ detectors and studies on ββ processes with INR-Kiev

Conclusion

1 • *Solid, well-known and evergreen detectors*

2 • *Widespread in many fields*

3 • *Improvements in performance and radiopurity achieved with time*

4 • *Technology constantly in development*

5
- *ULB inorganic crystal scintillators are powerful tools to investigate rare processes*
- *ULB NaI(Tl) has been succesfully used by DAMA/NaI and DAMA/LIBRA to investigate a particle Dark Matter component in the galactic halo by the annual modulation signature*

•Competitive on the frontier of investigation of rare processes