Crystal scintillators for low background measurements

RPSCINT2013
Kiev, September 16-20, 2013

R. Bernabei
University and INFN Roma Tor Vergata

Work with and legacy of Yuri

The ukrainian pioneer in Low Background Measurements.

Easy to work with, both because of scientific and human qualities

New limits on $2\beta^{+}$ decay processes in ^{106}Cd

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The first common work

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Search for double-β decay processes in ^{106}Cd with the help of a $^{106}CdWO_4$ crystal scintillator

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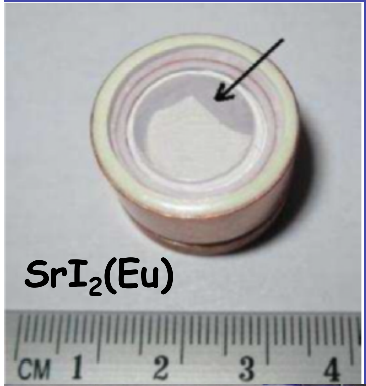
→ a living collaboration with his excellent group

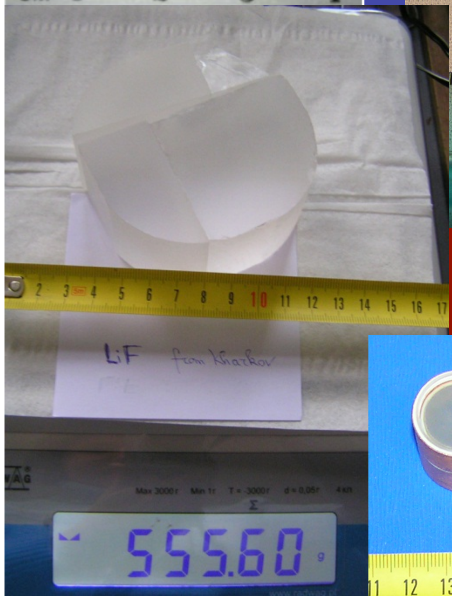
Memories of early days

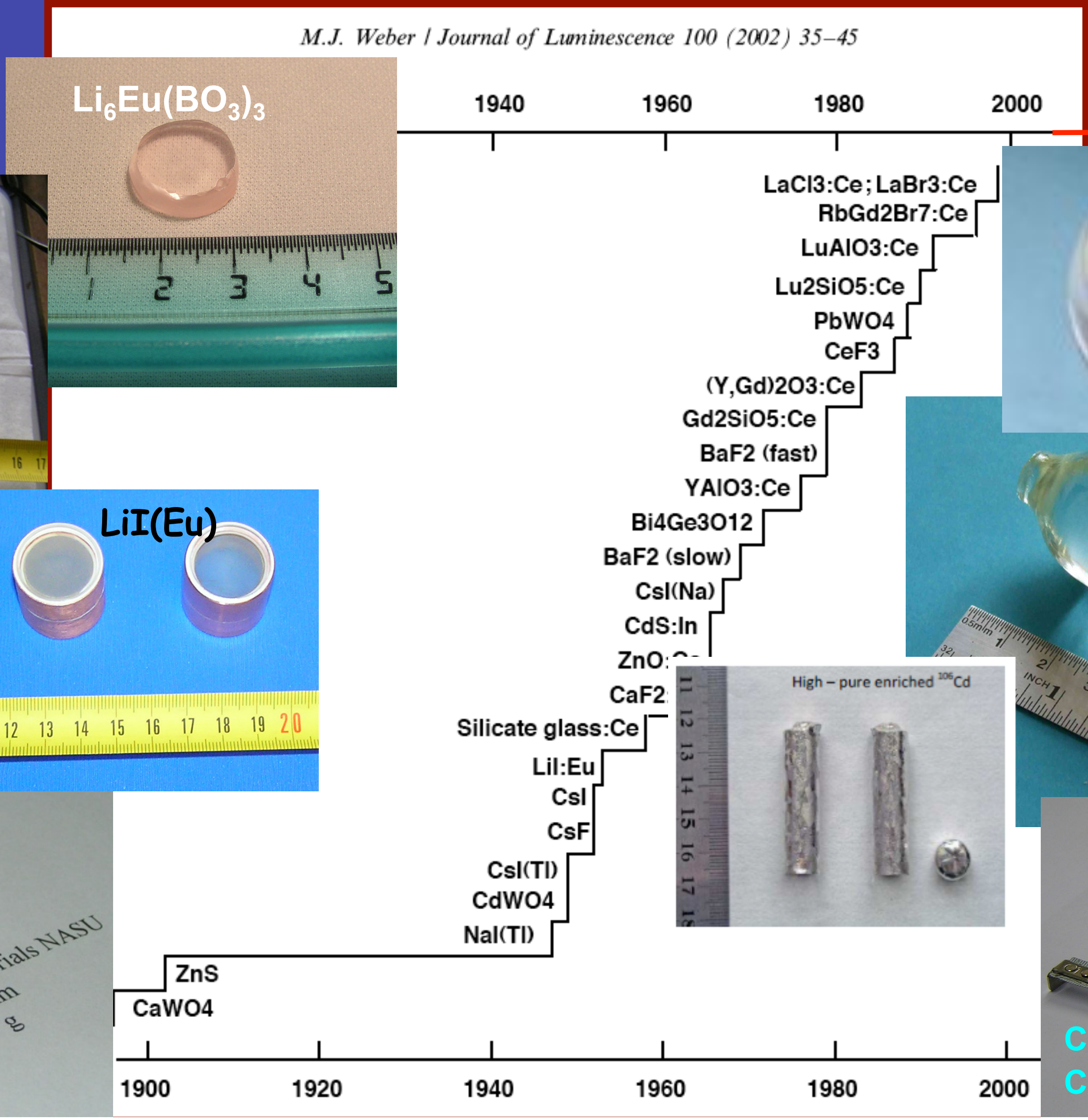
Continuing innovation

1903: Crookes used a ZnS-coated screen which scintillates when struck by α

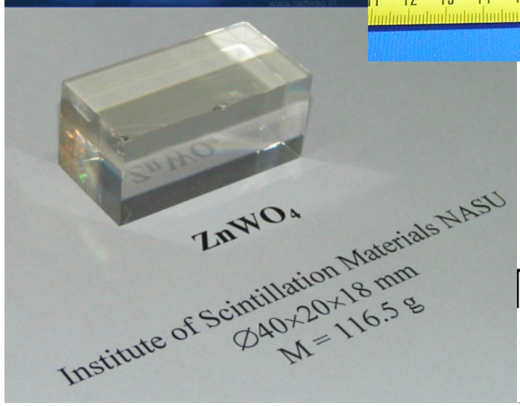
1944: Curran & Baker coated a PMT with ZnS → 1st scintillator that didn't require the human eye

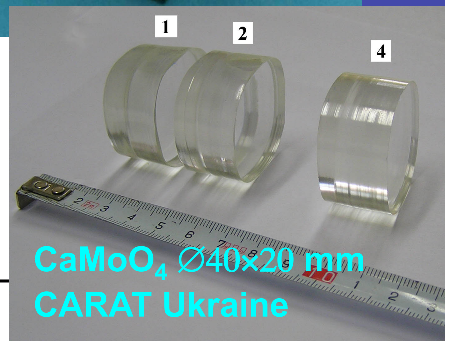




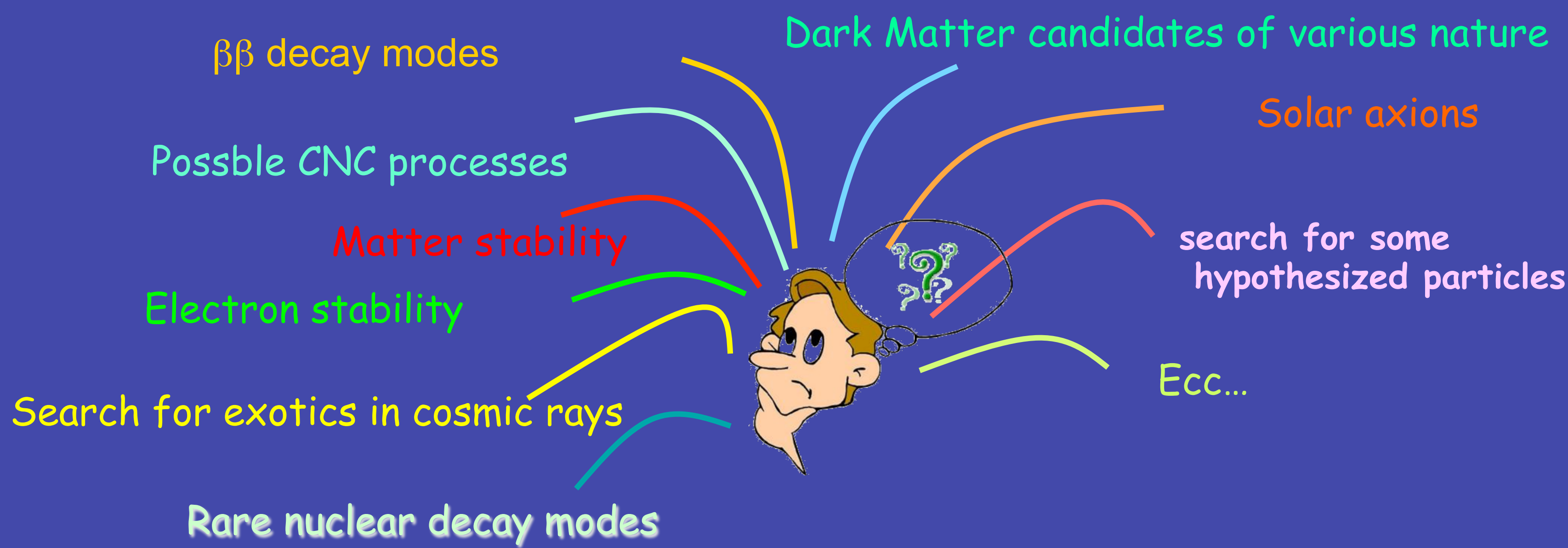


new developments





Old and new crystal scintillators in the investigations of rare processes: a leader role



Main requirements

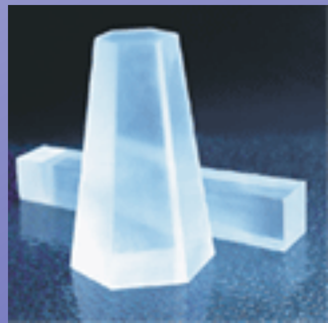
- ✓ Possibility of suitable radio-purity and mass (at least in perspective)
- ✓ Well known technology, acceptable cost, safety, high duty-cycle
- ✓ Excellent control and stability of all the running conditions feasible
- ✓ High detector performances: linearity of energy scale, high light response, …

But never forget:

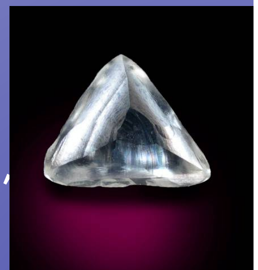
✧Detectors are just one component of a low background experiment

✧Each detector has its own features

✧Each ULB production cannot exactly be reproduced e.g. because of:



- change of sources of materials,
- unavoidability of materials from the same selection,
- possible activation/pollution in (long) storage at sea level,
- different additives, seeds
- different procedures due to time modification of the production equipments, of the safety rules,
- loss of some competences in the periodical change of involved people,



+

the highest is the initial radiopurity the most difficult is to avoid to lose it in the handling in the company, in the packing, in the handling in the experiment, etc.

In real low background

experiments

even a screw can make the

difference

To pursue the highest radiopurity always mandatory

<->

e.g. to overcome at most uncertainties

in Montecarlo background modeling and subtraction

(contaminants, limits, exact locations, etc.)

and to avoid heavy (and thus potentially uncertain)

data handling in complicate set-ups

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

…. contaminants

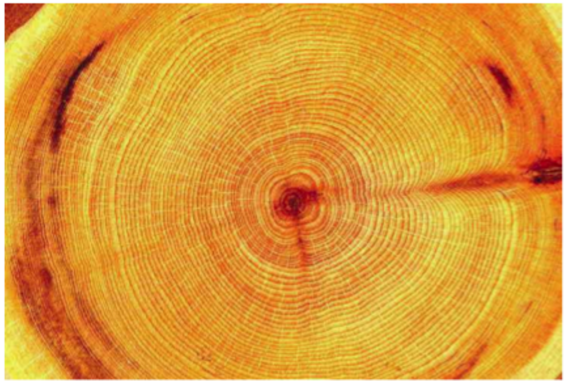
- ✓ Main sources of radioactive contamination of scintillation materials are naturally occurring radionuclides of ^{232}Th, ^{238}U, and ^{235}U families, and ^{40}K.
- ✓ The secular equilibrium of U/Th chains is generally broken in scintillation materials.
- ✓ Alpha active ^{147}Sm was detected in some scintillators at mBq/kg level.
- ✓ Then there are antropogenic ^{60}Co, ^{90}Sr-^{90}Y, ^{137}Cs nuclides.
- ✓ Some scintillation crystals consist of elements having radioactive isotopes, as e.g.: ^{152}Gd in GSO, ^{113}Cd in $CdWO_4$, ^{138}La in $LaCl_3$ and $LaBr_3$, ^{176}Lu in Lu_2SiO_5 and LuI_3.
- ✓ Cosmogenic radionuclides, i.e. created by high energy cosmic rays or/and by neutrons, were observed in scintillation materials:
  - ^{14}C in liquid scintillator,
  - ^{65}Zn in $ZnWO_4$,
  - ^{152}Eu in CaF_2(Eu),
  - ^{113m}Cd in $CdWO_4$.

  & ^{207}Bi in BGO whose origin is still not clear

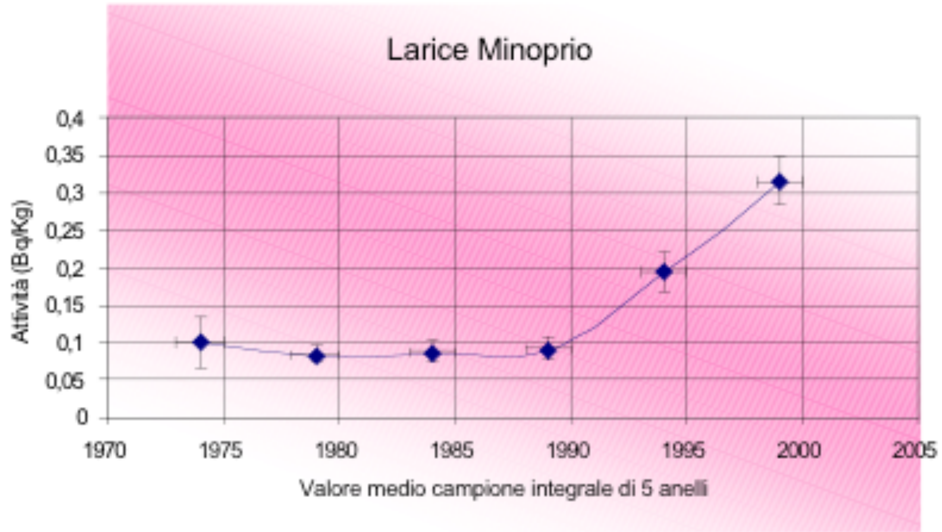
The highest sensitivity to measure internal contamination of crystal scintillators achieved in low bckg measurements where a scintillator operates as a detector:

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

A (Larix) in the Minoprio park



The corresponding ^{137}Cs activity as a function of the year

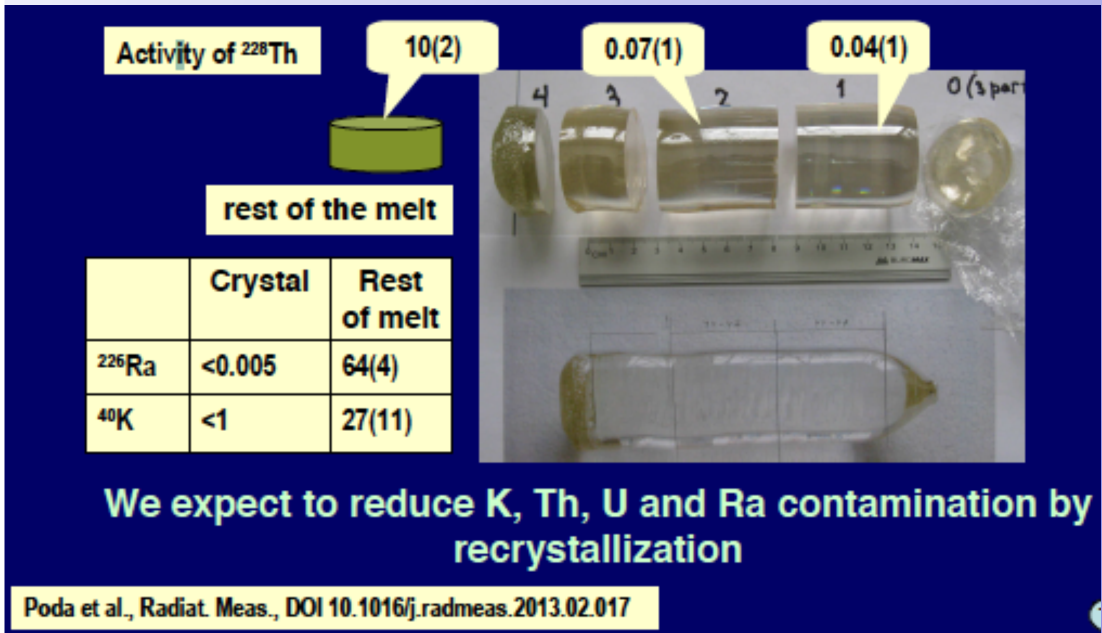


From a talk by E. Fiorini

Re-crystallization?

✓ Not effective for every kind of crystal

✓ Can be effective e.g. in $^{116}CdWO_4$ (to lower the internal Th)

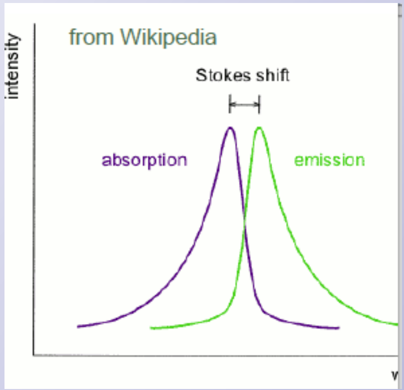


(activity in mB/kg)

from F. Danevich talk at MEDEX2013

✓ Risk of re-pollution in the whole procedure?

Main crystal scintillator characteristics:



Stokes shift

light yield

emission spectrum

decay time

density and Z

Afterglow (phosphorescence after some ms):

Caused by impurities or defects that create traps or metastable states with long lifetime → Pure crystals & e.g. with NaI(Tl) detectors in low bckg, blocking time can be used in the DAQ and rejection by PSD (well different time decay of scintillation pulse and afterglow single photoelectrons), etc.

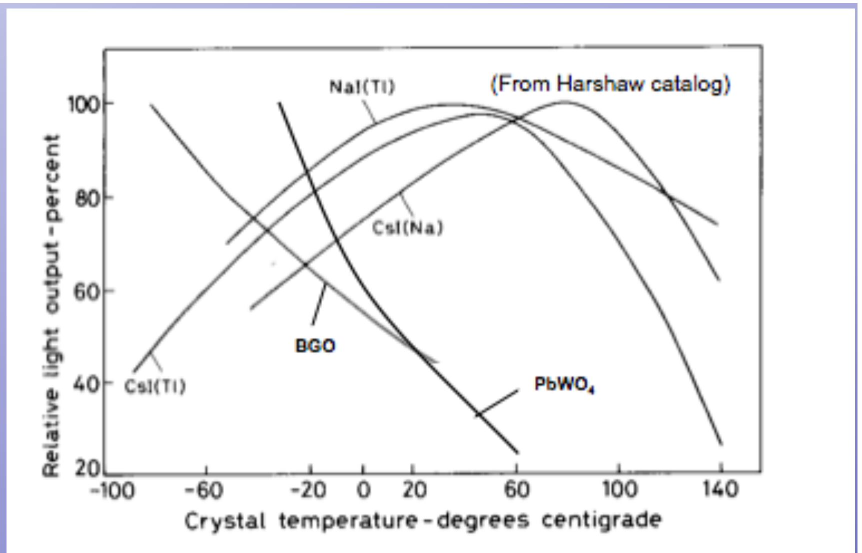
Properties of some inorganic scintillators

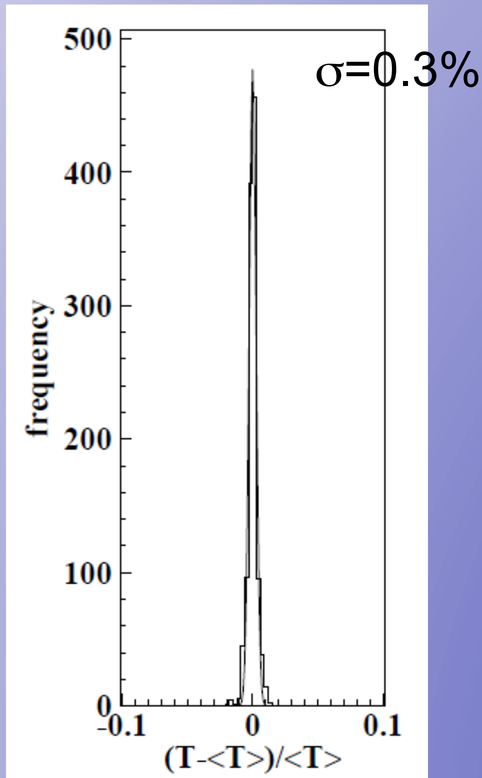
scintillator composition	density (g/cm^3)	index of refraction	wavelength of maximum emission (nm)	decay time constant (μs)	scintillation pulse height[1)]	notes
NaI	3.67	1.78	303	0.06	190	2)
NaI(Tl)	3.67	1.85	410	0.25	100	3)
CsI	4.51	1.80	310	0.01	6	3)
CsI(Tl)	4.51	1.80	565	1.0	45	3)
CaI(Na)	4.51	1.84	420	0.63	85	3)
KI(Tl)	3.13	1.71	410	0.24/2.5	24	3)
^{6}LiI(Eu)	4.06	1.96	470-485	1.4	35	3)
CaF_2(Eu)	3.19	1.44	435	0.9	50	
BaF_2	4.88	1.49	190/220 310	0.0006 0.63	5 15	
$Bi_4Ge_3O_{12}$	7.13	2.15	480	0.30	10	
$CaWO_4$	6.12	1.92	430	0.5/20	50	
$ZnWO_4$	7.87	2.2	480	5.0	26	
$CdWO_4$	7.90	2.3	540	5.0	40	
CsF	4.65	1.48	390	0.005	5	3)
CeF_3	6.16	1.68	300 340	0.005 0.020	5	
ZnS(Ag)	4.09	2.35	450	0.2	150	4)
GSO	6.71	1.9	440	0.060	20	
ZnO(Ga)	5.61	2.02	385	0.0004	40	4)
YSO	4.45	1.8	420	0.035	50	
YAP	5.50	1.9	370	0.030	40	

1) relative to NaI(Tl) 2) at 80 K 3) hygroscopic 4) polycrystalline

Temperature dependence of the light output of some inorganic crystals

e.g. in DAMA:
Continuous air conditioning double system + metallic housing of NaI(Tl) in direct contact with multi-ton metallic shield → huge heat capacity (≈10^6 cal/^{0}C) +
T continuously recorded + routine calibration each ≈10 days

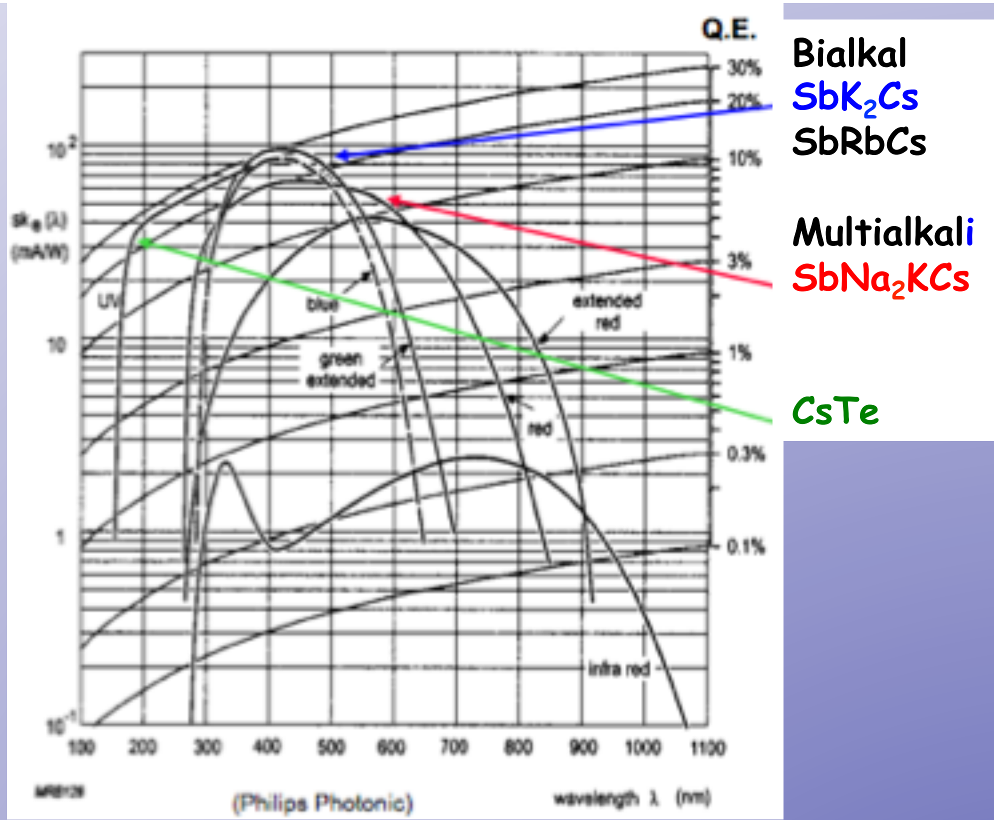




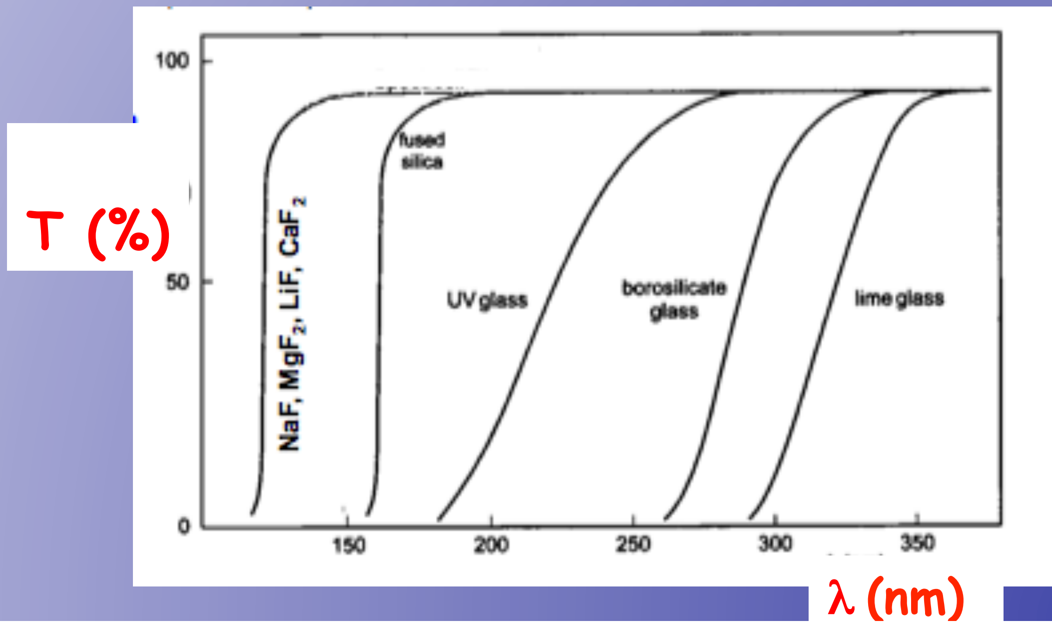
Distribution of the relative variations of the operating T of the detectors in the whole DAMA/LIBRA-phase1 (7 annual cycles)

It can further limits Sensitivity of PSD at keV region

Q.E. of typical photocathodes



Transmission of PMT windows

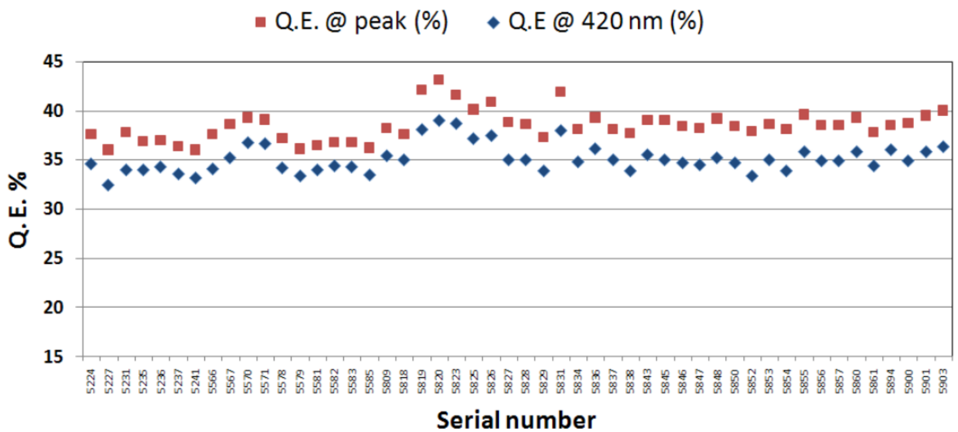


A recent development

carried out by HAMAMATSU co. in order to produce high Q.E. PMTs with size, radiopurity, gain, dark current, etc. according to the requirements of the DAMA/LIBRA experiment

→ DAMA/LIBRA-phase2

J. Instrum. 7, P03009 (2012).



Average values:

Q.E. @ peak 38.5% (1.6% RMS)

Q.E. @ λ=420 nm 35.1 % (1.4% RMS)

Best value:

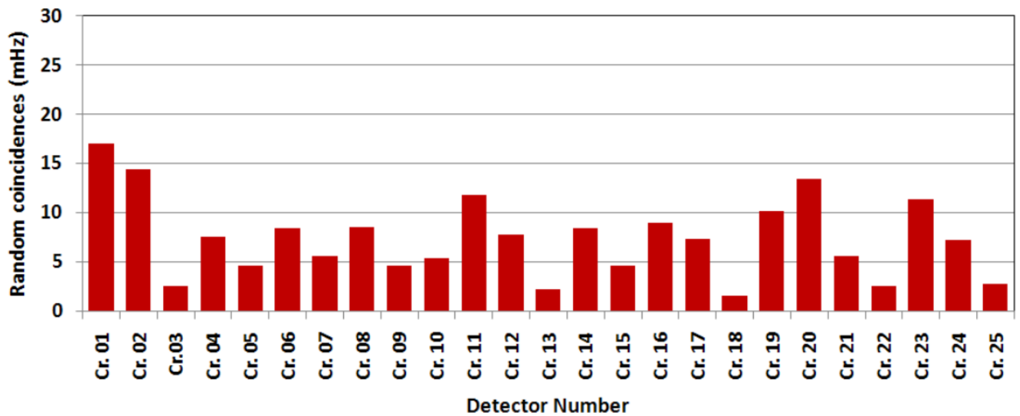
39% @ 420 nm & 43.1% @ peak

Worst value:

32.5% @ 420 nm & 36% @ peak

Random coincidences between the 2 PMTs of each crystal (here Δt=100 ns):

→ typically <<0.02 Hz



Radiopurity of crystal scintillators

Radioactive contamination of crystal scintillators (mBq/kg)

arXiv:0903.1539[nucl-ex]

Scintillator	Total α activity (U + Th)	^{228}Th	^{226}Ra	^{40}K	Particular 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)		0.002	0.008		6 (^{134}Cs) 14 (^{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: $T_{1/2} = 3.78\times10^{10}$ y, its isotopic abundance (2.59%) [25], and chemical formula of the LuI_3 compound.

Annotations (arrows to table rows): DAMA+INR Kiev ($ZnWO_4$); INR Kiev & DAMA+INR Kiev ($CdWO_4$); DAMA/LIBRA (NaI(Tl), 0.08 row); KIMS (CsI(Tl)); DAMA (CaF_2(Eu)); ELEGANT VI (CaF_2(Eu), second row)

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

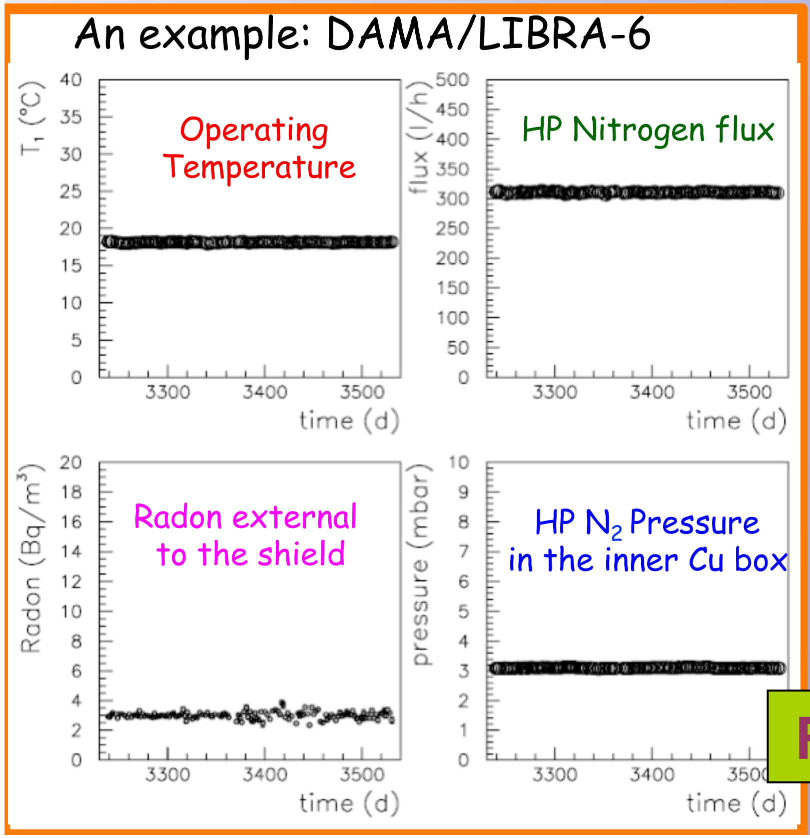
- Time-amplitude analysis
- Pulse-shape discrimination
- Energy spectra analysis

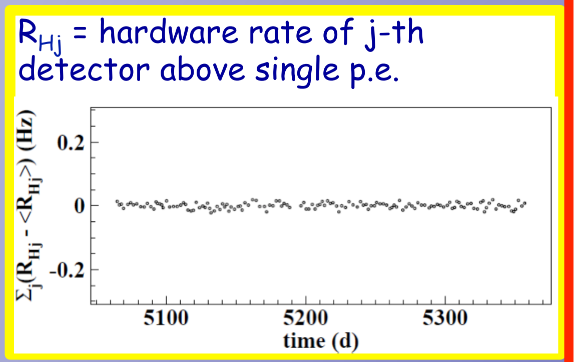
STABILITY

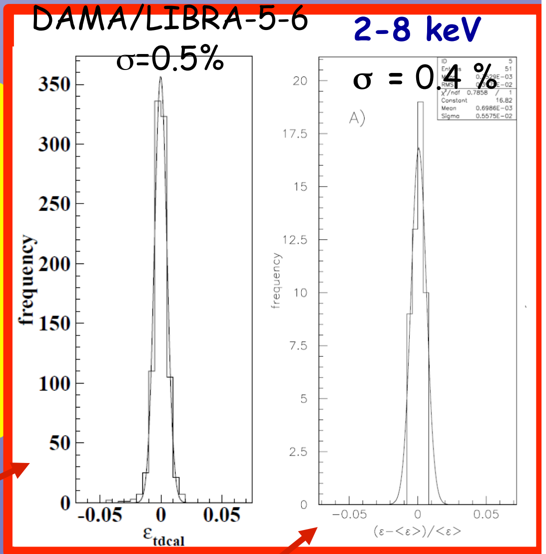
Crystal scintillators allow:

1) **well controlled operational conditions and monitoring;**
2) **high reproducibility, high stability, etc. (they do not require re-purification or cooling down/warming up procedures);**
3) **high duty cycle.**

Stability of the running conditions can be continuously monitored along all the data taking







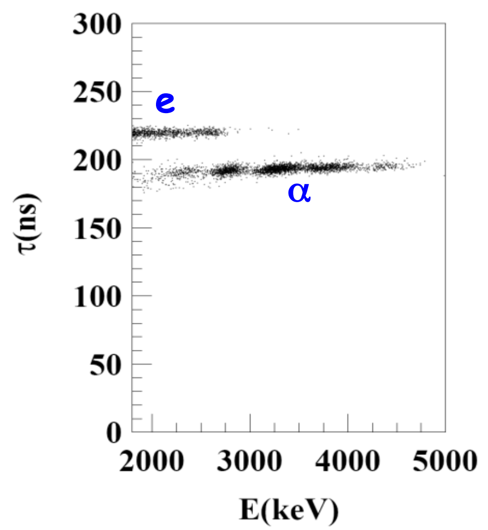
...and more:

the energy scale, the efficiencies

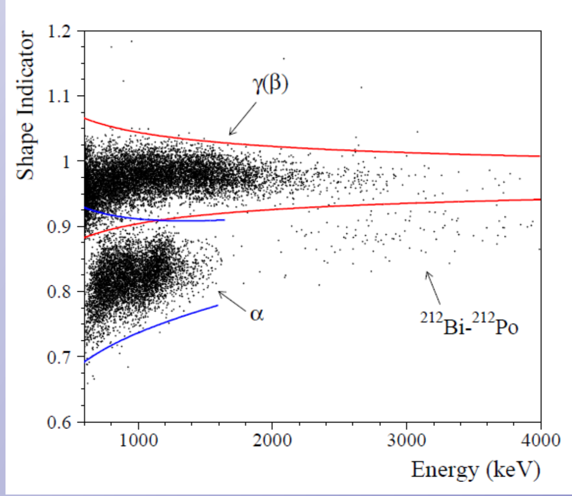
Running conditions stable at level < 1%

PSD

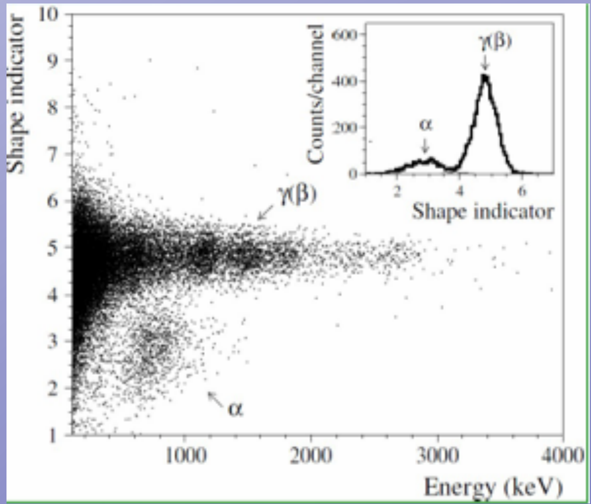
✓ Very good or ideal in the alpha region, e.g.:



NaI(Tl)



$^{116}CdWO_4$

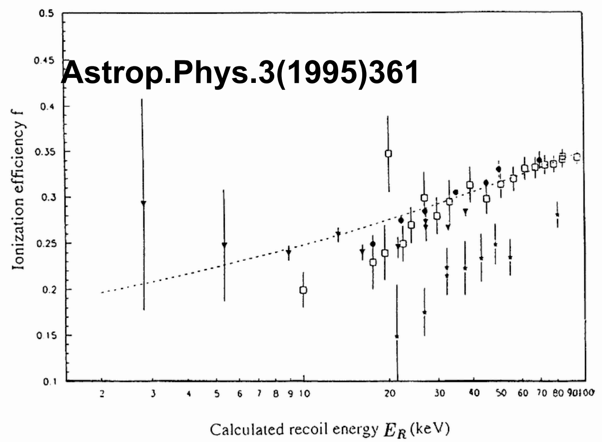


$ZnWO_4$

✓ It degrades for every scintillators when lowering the energy down to become marginal in keV region

QUENCHING FACTORs in keV region

Ex. of different *q* determinations for Ge



differences are generally 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 and on the impurities/trace contaminants; in LXe e.g. on trace impurities, on initial UHV, on presence of degassing/releasing materials in the Xe, on thermodynamical conditions, on possibly applied electric field, etc.

… 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)).

Other interesting arguments in arXiv:0911.3041[nucl-ex] → see talk by Tretyak

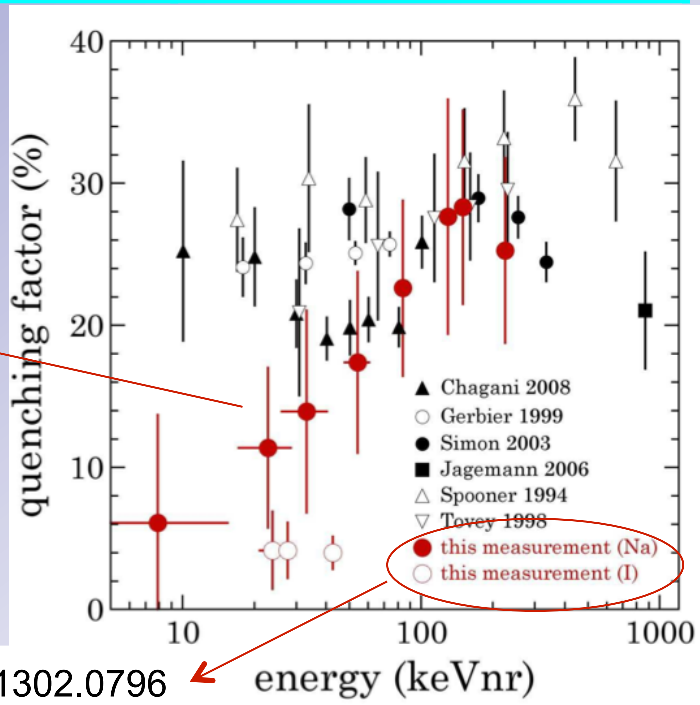
direct measurements of q. f. are performed with reference detectors that in some cases have features quite different from the detectors used in the running conditions.
The nature of these measurements, the parameterisation, the used neutron beam/ sources may not point out all the possible contributions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

Further uncertainties in recoils' quenching

EPJC53(2008)205

- ***In crystals, ions move in a different manner than that in amorphous materials.***
- In the case of motion along crystallographic axes and planes, a channeling effect is possible, which is manifested in an anomalously deep penetration of ions into the target.

Channeling effect in crystals

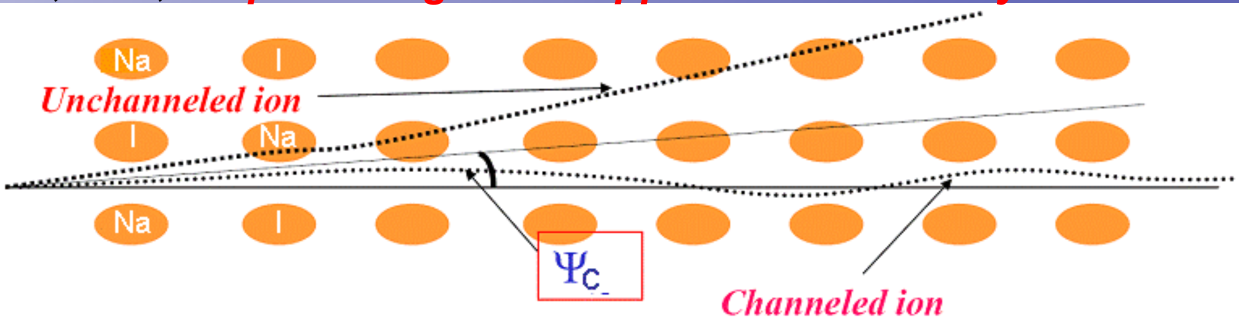
Well-known effect, discovered on 1957, when a deep penetration of $^{134}Cs^{+}$ ions into a Ge crystal to a depth $\lambda_c \approx 10^3$ Å was measured (according to SRIM, a 4 keV Cs^+ ion would penetrate into amorphous Ge to a depth λ_a = 44 Å, S_n/S_e = 32 and q=0.03). Within a channel, mostly electronic stopping takes place (in the given example, $\lambda_c \approx \lambda_a/q \approx$ 1450 Å).

- Occurs in **crystalline** materials due to correlated collisions of ions with target atoms.
- Steering of the ions through the open channels can result in **ranges several times the maximum range** in no-steering directions or in **amorphous materials**.
- **Electronic losses** determine the range and there is very little straggling.
- When a low-energy ion goes into a channel, its energy losses are mainly due to the electronic contributions. This implies that a channeled ion transfers its energy mainly to electrons rather than to the nuclei in the lattice and, thus, its ***quenching factor approaches the unity***.

$$R_{ion}(E) \approx R_{el.}(E)$$

$$L_{ion} \approx L_{el}$$

$$q(E) \approx 1$$



Example: neutron calibrations of NaI(Tl) detectors

EPJC53(2008)205

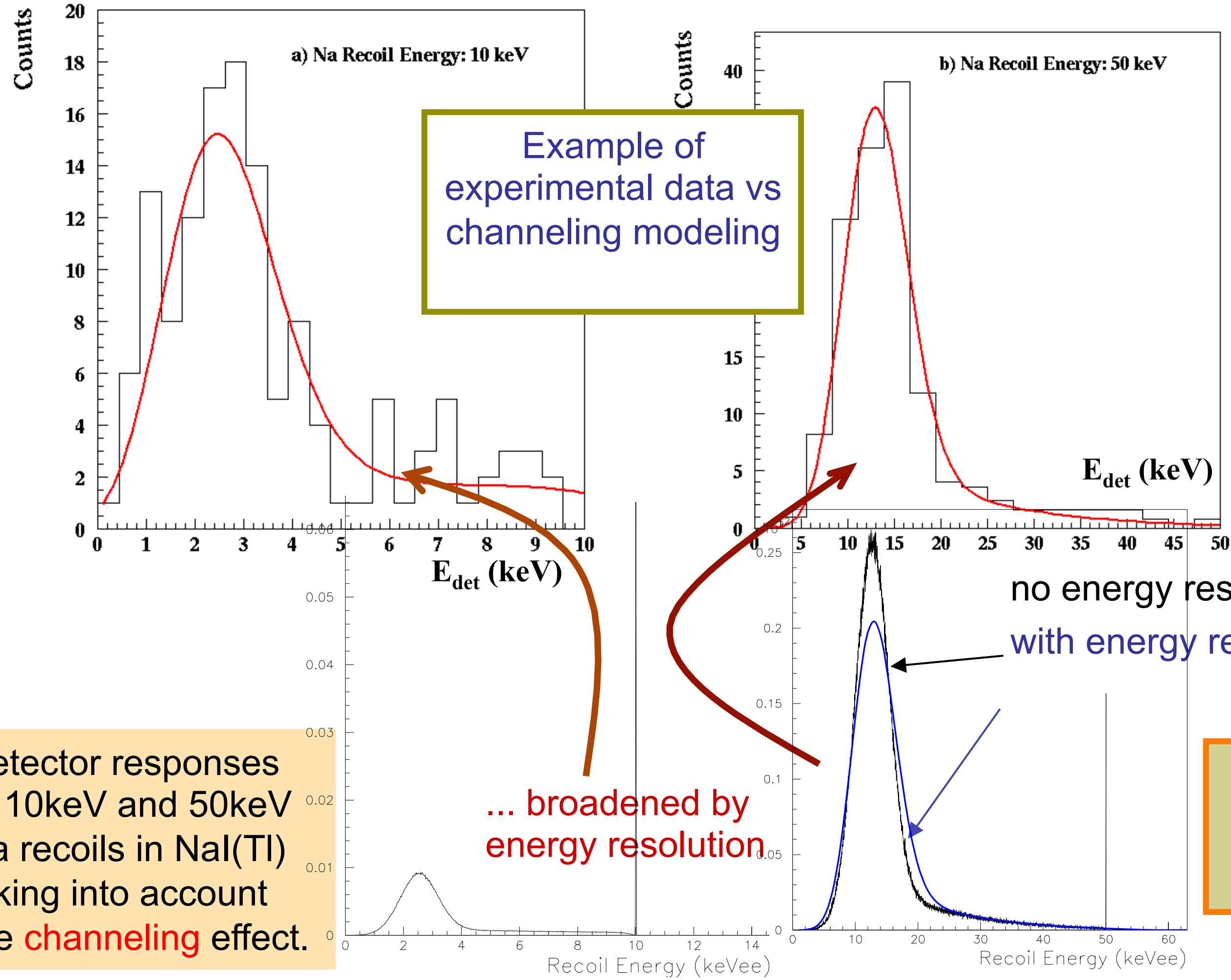
MEASUREMENT OF THE SCINTILLATION EFFICIENCY OF Na RECOILS IN NaI(Tl) DOWN TO 10 keV NUCLEAR RECOIL ENERGY RELEVANT TO DARK MATTER SEARCHES

H. CHAGANI*, P. MAJEWSKI**, E. J. DAW, V. A. KUDRYAVTSEV, and N. J. C. SPOONER

arXiv:physics/0611156 (IDM 2006)

SICANE: a Detector Array for the Measurement of Nuclear Recoil Quenching Factors using a Monoenergetic Neutron Beam

NIMA 507 (2003) 643



Detector responses to 10keV and 50keV Na recoils in NaI(Tl) taking into account the channeling effect.

- neutron data can contain channeled events
- but – owing to the low-statistics of these measurements and to the small effect looked for – they cannot be identified
- E.g. at higher energy and for Iodine recoils the channeling effect becomes less important and gives more suppressed contributions in the neutron scattering data

Therefore, there is no hope to identify the channeling effect in existing neutron data

Some alternative channeling models with respect to EPJC53(2008)205:

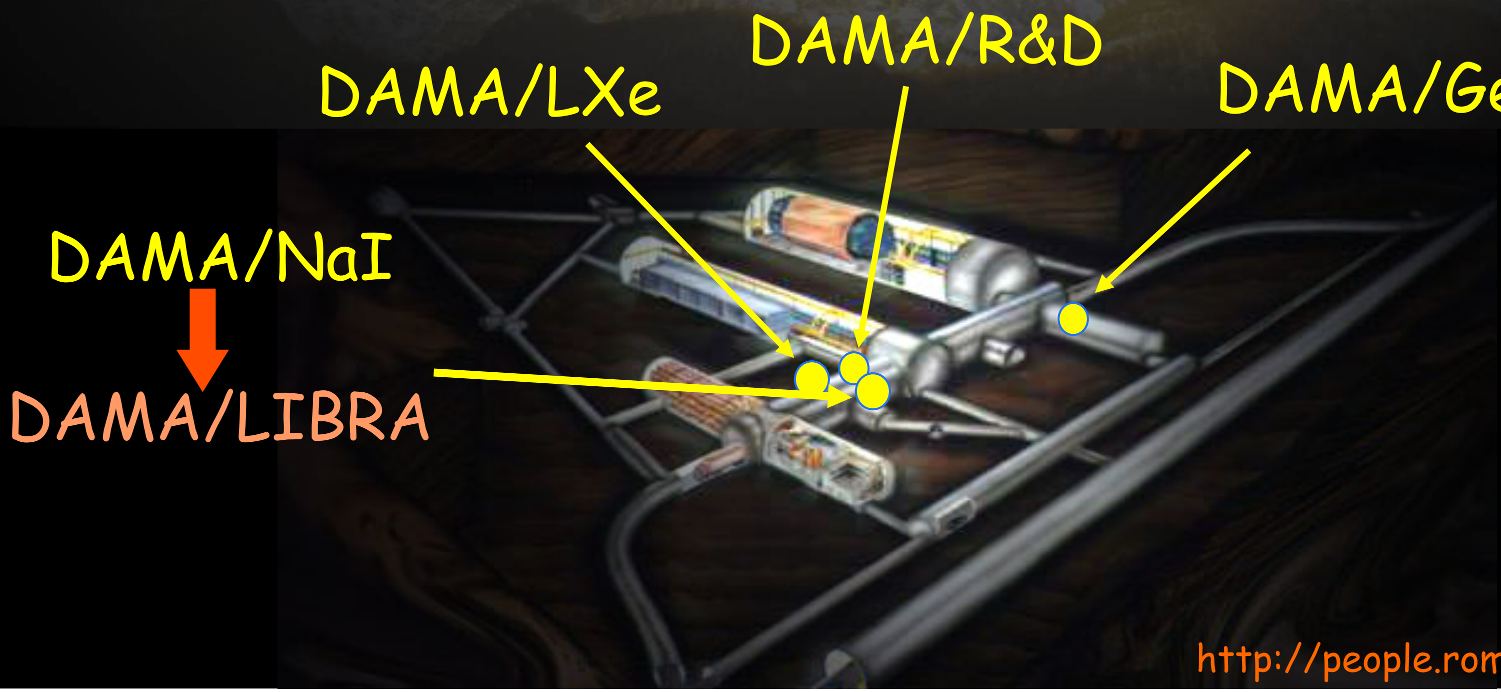
- ✓ larger probabilities of the planar channeling expected in Tech. Phys. 53, 1578 (2008).

- ✓ analytical calculation in Astropart. Phys. 11, 19 (2010) claiming that the channeling effect holds for nuclear recoils coming from outside a crystal and not from nuclear recoils produced inside it, due to the blocking effect; nevertheless, although some amount of blocking effect may be present, the precise description of the crystal lattice with dopant and trace contaminants is quite difficult and analytical calculations require some simplifications which affect the result.

Roma2,Roma1,LNGS,IHEP/Beijing

+ by-products, small scale expts. and several detectors developments: INR-Kiev

Many low background crystal scintillators developed and used



http://people.roma2.infn.it/dama

20 years of developments in highly radiopure NaI(Tl)
Harshaw, Quartz&Silice, Crismatec, Saint Gobain





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

(detector installation in HP Nitrogen atmosphere)


DAMA/LIBRA setup: NIMA592(2008)297
See also references therein
upgrading on September 2008

Which NaI(Tl) ?

N.B.: to get ^{40}K multiply by 10^{-4}

Qualification of NaI(Tl)	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 (CL, BL) + Kyropoulos growth
Ultra Low Background (project Gran Sasso)	<< 40	< 5	< 5	Purified raw materials NaI and TlI + Crafted Kyropoulos growth + Handling protocol

SAINT-GOBAIN CRYSTALS

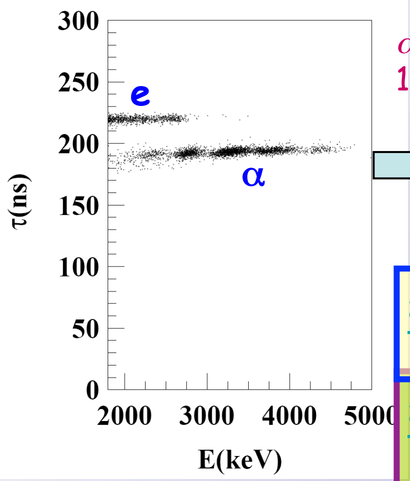
+ as known, determination with the highest sensitivity by measurements with the detectors deep underground:

DAMA/LIBRA see NIMA592(2008)297;
former DAMA/NaI see NCIMA112(1999)545, EPJC18(2000)283 …

1) Not all the cookings have the same taste

2) As whatever VLB or ULB detector used in whatever search for rare processes, VLB or ULB NaI(Tl) cannot be simply "bought"

Some on residual contaminants in new 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

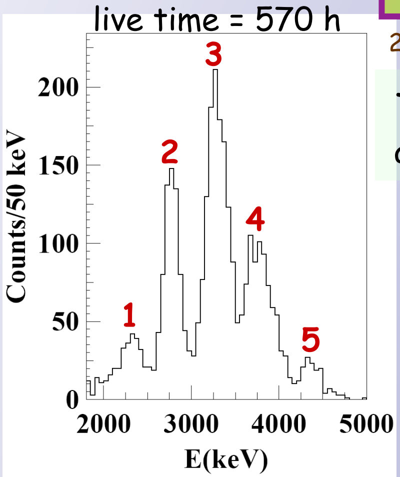
Second generation R&D for new DAMA/LIBRA crystals: new selected powders, physical/chemical radiopurification, new selection of overall materials, new protocol for growing and handling

^{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



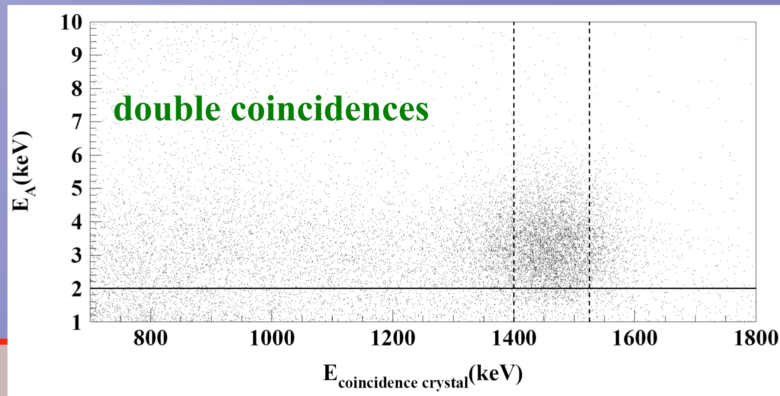
^{238}U chain splitted into 5 subchains: $^{238}U \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{210}Pb \rightarrow {}^{206}Pb$

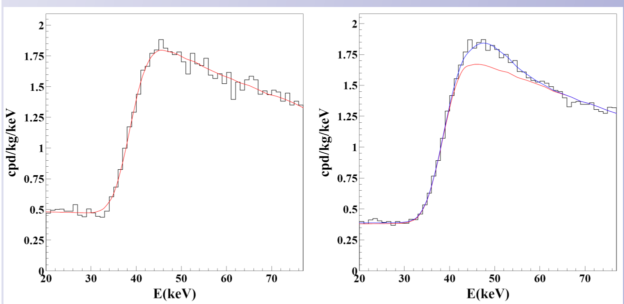
Thus, in this case: (2.1±0.1) ppt of ^{232}Th; (0.35 ±0.06) ppt for ^{238}U

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 (mean value 13 ppb)





^{129}I and ^{210}Pb

^{129}I/^{nat}I ≈1.7×10^{-13} for all the new detectors

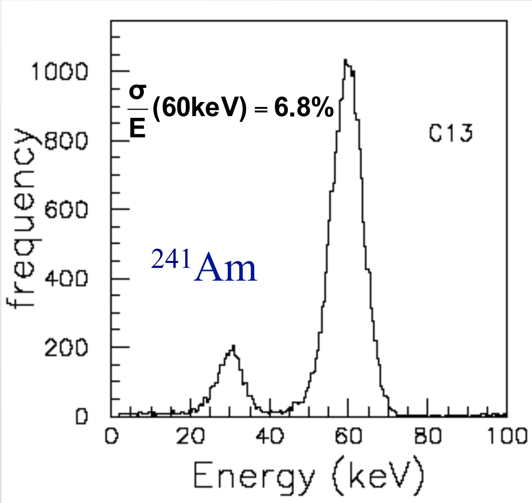
^{210}Pb in the new detectors: (5 – 30) μBq/kg.

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

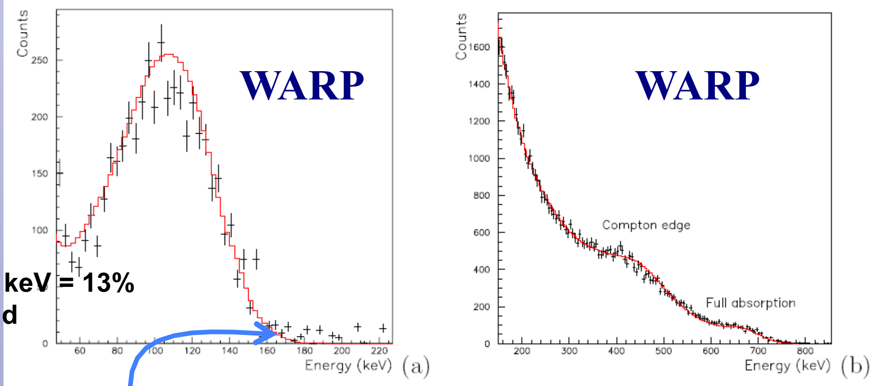
For details and other information see NIMA592(2008)297

Examples of energy resolutions

DAMA/LIBRA ULB NaI(Tl)



NIMA 574 (2007) 83



σ/E @ 122 keV = 13%
at zero field

Fig. 2. Energy spectra taken with external γ-ray sources, superimposed with the corresponding Monte Carlo simulations. (a) ^{57}Co source ($E = 122$ keV, B.R. 85.6%, and 136 keV, B.R. 10.7%), (b) ^{137}Cs source ($E = 662$ keV).

subtraction of the spectrum ?

ZEPLIN-II

AP 28 (2007) 287

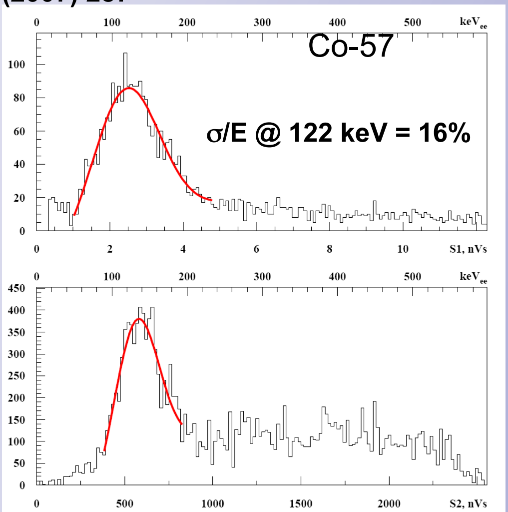


Fig. 5. Typical energy spectra for ^{57}Co γ-ray calibrations, showing S1 spectrum (upper) and S2 spectrum (lower). The fits are double Gaussian fits which incorporate both the 122 keV and 136 keV lines in the ^{57}Co γ-ray spectrum. The energy resolution of the detector is derived from the width of the S1 peak, coupled with calibration measurements at other line energies.

XENON10 XENON10

σ/E @ 122 keV = 17%

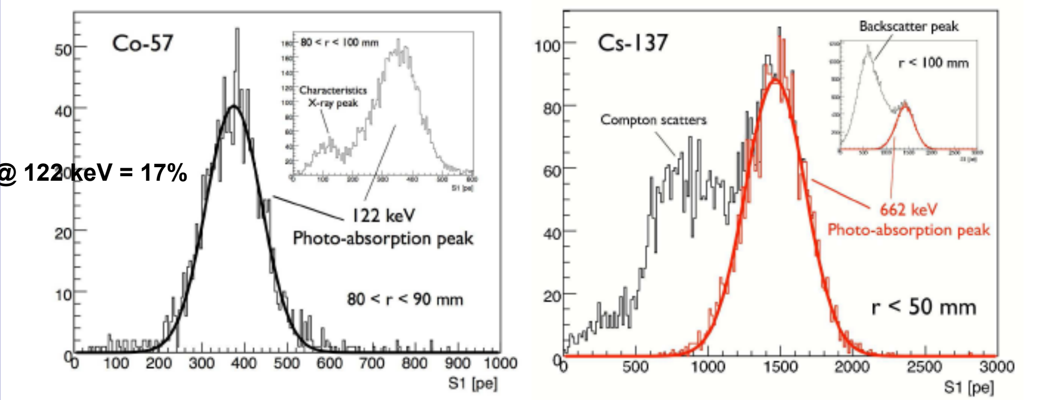
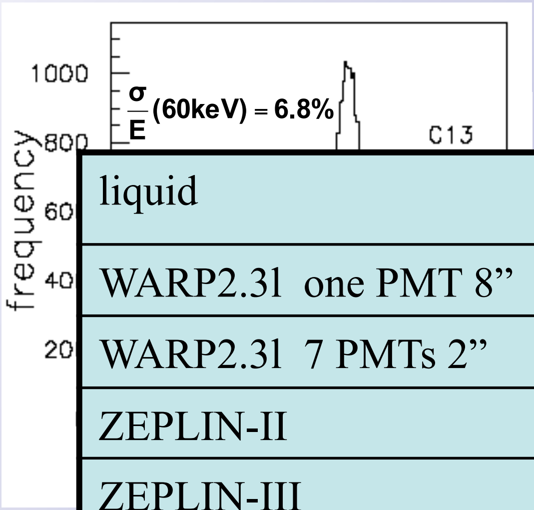


Figure 3. (left) S1 scintillation spectrum from a ^{57}Co calibration. The light yield for the 122 keV photo-absorption peak is 3.1 p.e./keV. (right) S1 scintillation spectrum from a ^{137}Cs calibration. The light yield for the 662 keV photo-absorption peak is 2.2 p.e./keV.

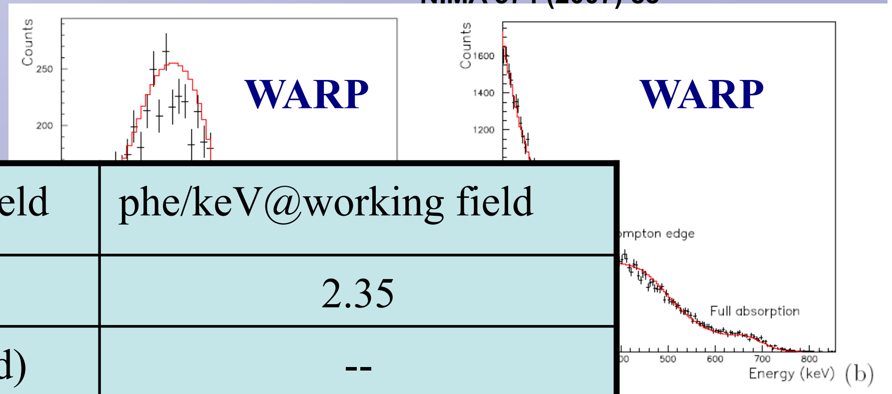
JoP: Conf. Ser. 65 (2007) 012015

Examples of energy resolutions

DAMA/LIBRA ULB NaI(Tl)



NIMA 574 (2007) 83



perimposed with the
122 keV, B.R. 85.6%,

rum ?

liquid	phe/keV@zero field	phe/keV@working field
WARP2.31 one PMT 8”	--	2.35
WARP2.31 7 PMTs 2”	0.5-1 (deduced)	--
ZEPLIN-II	1.1	0.55
ZEPLIN-III		1.8
XENON10	--	2.2 (^{137}Cs), 3.1 (^{57}Co)
XENON100	2.7	1.57 (^{137}Cs), 2.2 (^{57}Co)
Neon	0.93	field not foreseen

DAMA/LIBRA-phase1 : 5.5 – 7.5 phe/keV

AP 28 (2007) 287

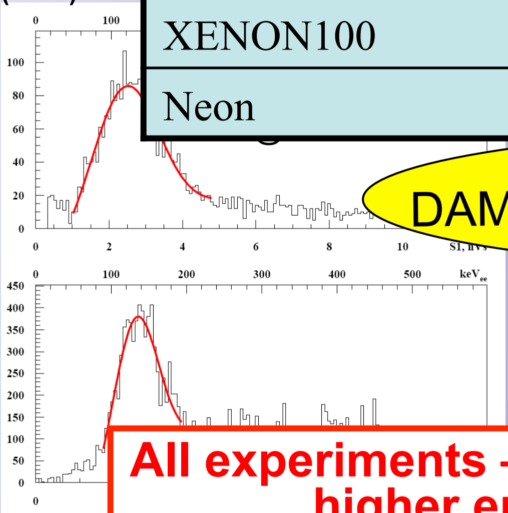
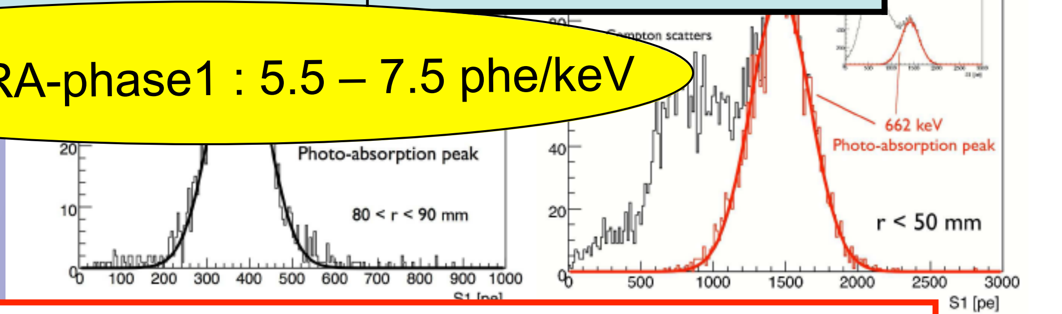


Fig. 5. Typical energy ... (upper) and S2 spectrum (lower). The fits are double Gaussian fits which incorporate both the 122 keV and 136 keV lines in the ^{57}Co γ-ray spectrum. The energy resolution of the detector is derived from the width of the S1 peak, coupled with calibration measurements at other line energies.



All experiments – except DAMA – use only calibration points at higher energy with extrapolation to low energy

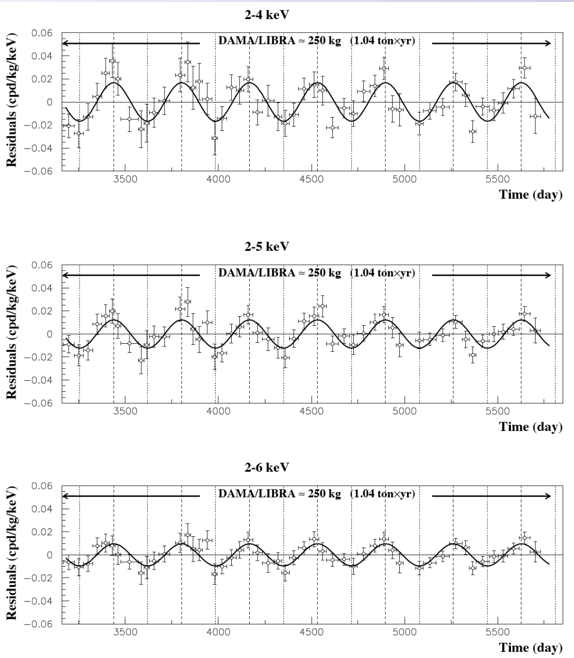
...he 122 keV ...ation. The light yield for the 662 keV photo-absorption peak is 2.2 p.e./keV.

JoP: Conf. Ser. 65 (2007) 012015

The case of DAMA/LIBRA

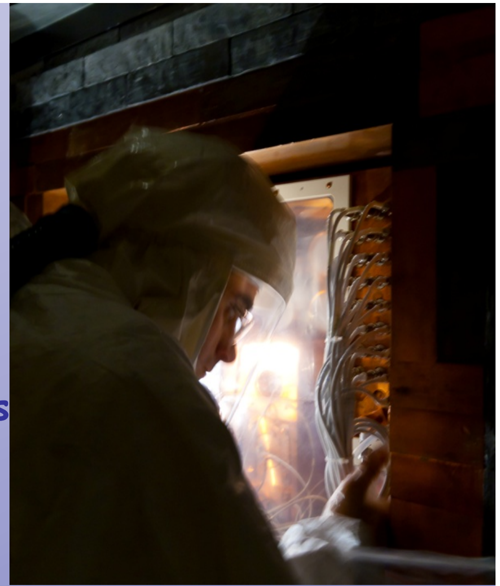
Phase1 concluded

9.3 σ C.L: DM model independent evidence cumulatively with DAMA/NaI
(EPJC2008,EPJC2010,arXiv:1308.5109,..)



Phase2 running

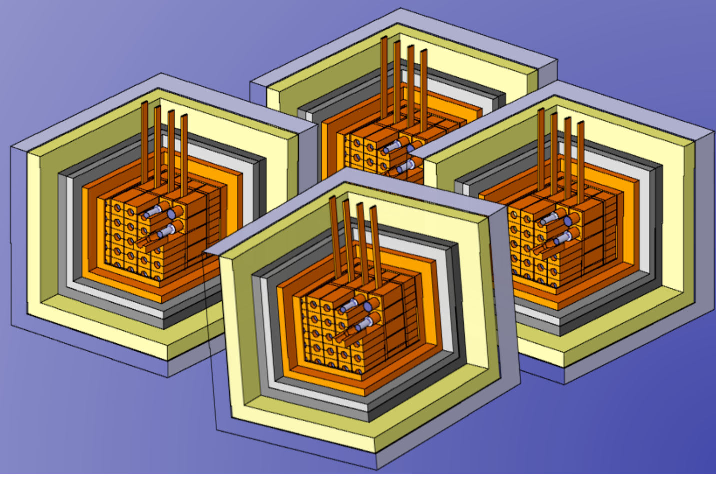
- Suitable exposure planned in the new configuration to deeper study the nature of the particles and features of related astrophysical, nuclear and particle physics aspects
- New investigation on dark matter peculiarities and second order effects
- Special data taking for other rare processes



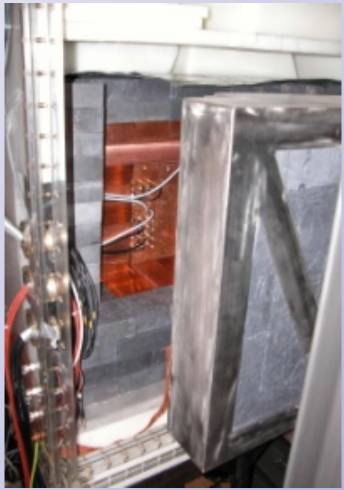
→ further implementations for the future

→ DAMA/1ton proposed since 1996

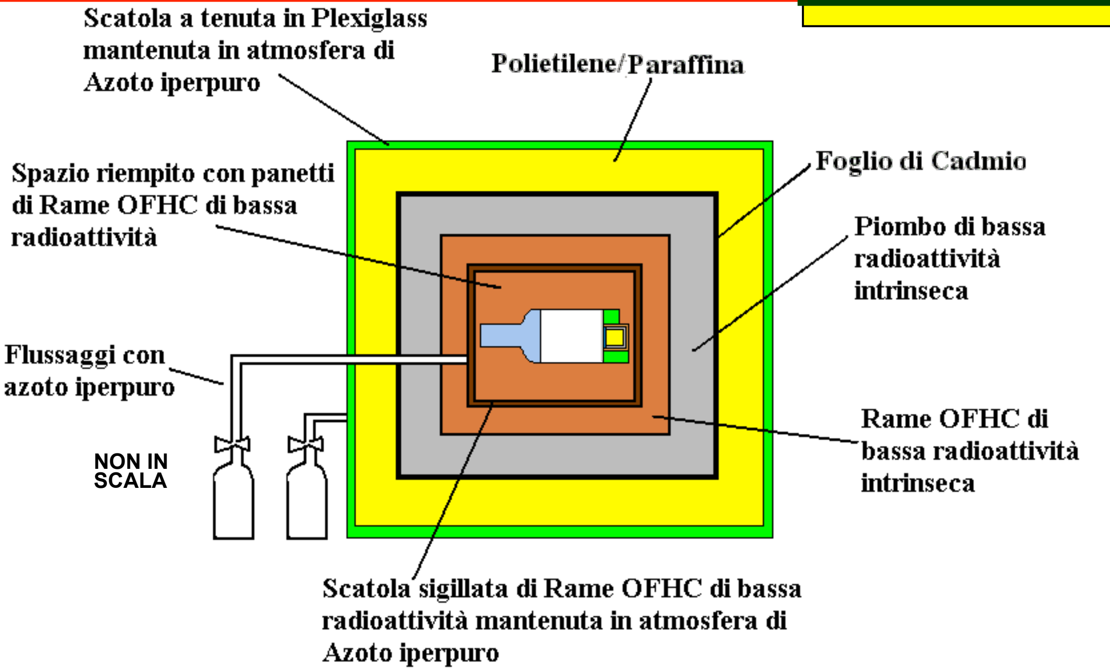
Original design consists in adding other 3 replicas of DAMA/LIBRA in 3 similar installations



DAMA/R&D



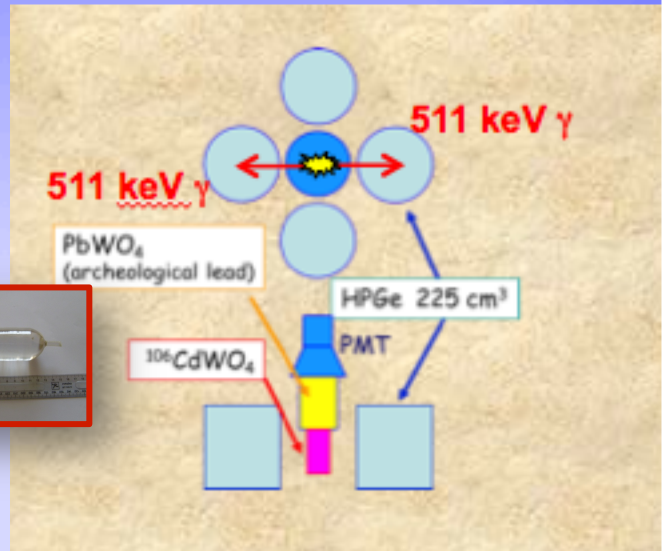
Schema of the setup

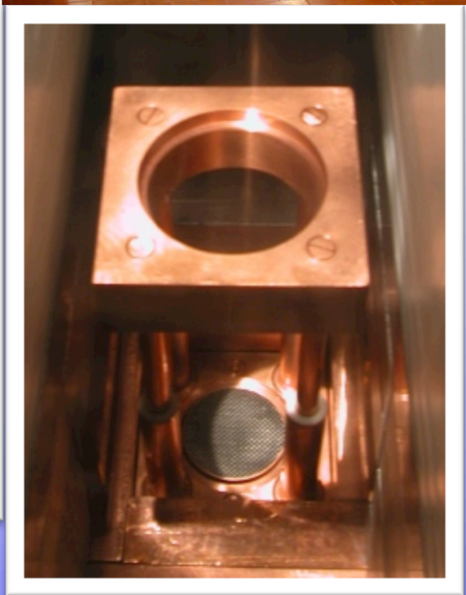


Radio-purity of the materials (95%C.L.):

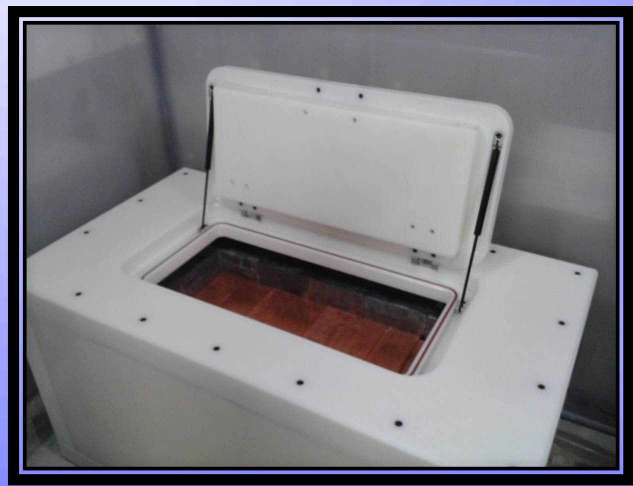
Materiale	^{238}U (ppb)	^{232}Th (ppb)	^{nat}K (ppm)
Cu	< 0.5	< 1	< 0.6
Pb boliden	< 8	< 0.03	< 0.06
Pb boliden2	< 3.6	< 0.027	< 0.06
Polish Pb	< 7.4	< 0.042	< 0.03
Polietilene	< 0.3	< 0.7	< 2
Plexiglass	< 0.64	< 27.2	< 3.3

DAMA/Ge and LNGS STELLA facility



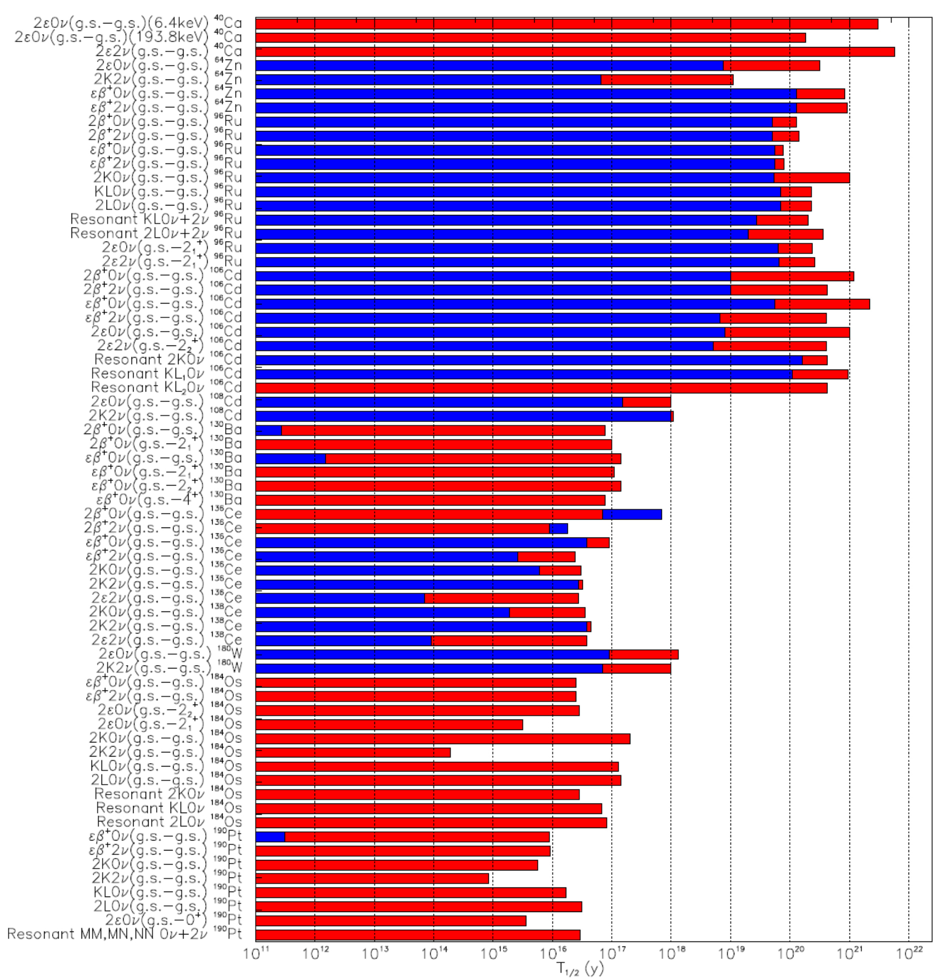


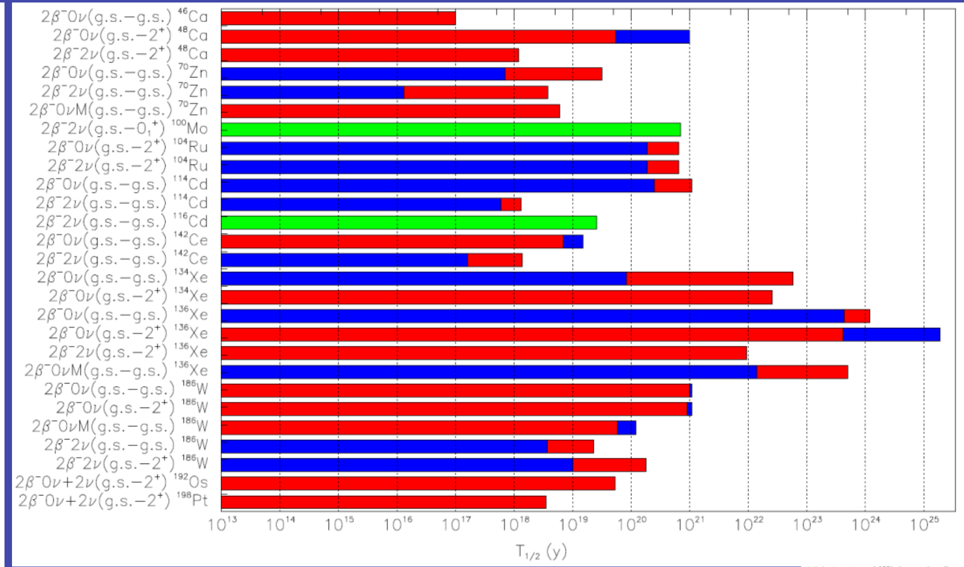
DAMA/CRYS



During installation

Summary of searches for ββ decay modes (partial list)





ARMONIA: New observation (green) of $2\nu2\beta^-$ $^{100}Mo \rightarrow ^{100}Ru$ (g.s.→0_1^+) decay
NPA846 (2010)143

AURORA: New observation of $2\nu2\beta^-$ ^{116}Cd decay
NPAE2012

- Many competitive limits obtained on lifetime of $2\beta^+$, $\varepsilon\beta^+$ and 2ε processes (^{40}Ca, ^{64}Zn, ^{96}Ru, ^{106}Cd, ^{108}Cd, ^{130}Ba, ^{136}Ce, ^{138}Ce, ^{180}W, ^{190}Pt, …).
- First searches for resonant ββ decays in some isotopes

$T_{1/2}$ experimental limits by DAMA (in red) and previous ones (in blue). All the limits are at 90% C.L. except for $0\nu2\beta^+$ in ^{136}Ce and $2\beta^-0\nu$ in ^{142}Ce at 68% C.L.. In green observed

Many publications on detectors developments and results
Many future measurements in preparation

Other scintillators by DAMA and by DAMA+INR-Kiev mainly to investigate ββ decay modes with source=detector approach

CaF_2(Eu)	Bicron/Crismatec(Saint Gobain)	NPA 789 (2007) 15 NPA 705 (2002) 29 NPB 563 (1999) 97
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 to appear
LiF(W)	Ukraine	NPA 806 (2008) 388
^{7}LiI(EU)	Ukraine	NIM704(2013) 40
$ZnWO_4$	Ukraine	JPG: NPP 38(2011) 115107 NIMA626(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	JPG: NPP 38(2011) 015103 NPA 824 (2009) 101
Li_2MoO_4	Ukraine	NIMA 607 (2009) 573
$Li_6Eu(BO_3)_3$	Ukraine	NIM A572 (2007) 734
SrI_2	Ukraine	NIMA670 (2012) 10
$CdWO_4$	Ukraine dev.towards	EPJA 36 (2008), 167 PRC 76 (2007) 064603
$^{106}CdWO_4$	Ukraine	PRC85(2012)044610, NIMA 615 (2010) 301 in measurement in STELLA
$^{116}CdWO_4$	Ukraine	JINS6(2011) 08011, in measurement in DAMA/R&D

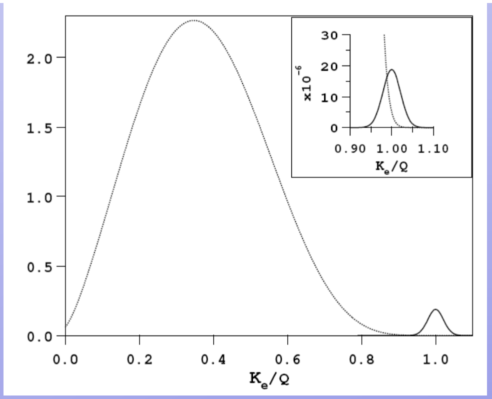
and also polycrystalline powder.

ZnS(Ag)	Saint-Gobain	MPLA 27, No. 8 (2012) 1250031

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

Isotope	Nat. Ab. (%)	Q (keV)	Decay Mode	Scintillator
^{64}Zn	48.63	1095.7	$\varepsilon\beta^+, 2\varepsilon$	$ZnWO_4$, $CdWO_4$
^{70}Zn	0.62	998.5	$2\beta^+$	$ZnWO_4$, $CdWO_4$
^{180}W	0.12	144	2ε	$ZnWO_4$, $CdWO_4$, $PbWO_4$
^{186}W	28.43	489.9	$2\beta^-$	$ZnWO_4$, $CdWO_4$, $PbWO_4$
^{106}Cd	1.25	2771	$2\beta^+$, $\varepsilon\beta^+$	$^{106}CdWO_4$
^{108}Cd	0.89	269	2ε	$CdWO_4$
^{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	2ε	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	$2\beta^+$, $\varepsilon\beta^+$	$CeCl_3$, CeF_3, $CeBr_3$
^{138}Ce	0.251	693	2ε	$CeCl_3$, CeF_3, $CeBr_3$
^{142}Ce	11.114	1416.9	$2\beta^-$	$CeCl_3$, CeF_3, $CeBr_3$
^{130}Ba	0.106	2611	$2\beta^+$, $\varepsilon\beta^+$, 2ε	BaF_2, $BaCl_2(Eu)$, $BaI_2(Eu)$
^{92}Mo	14.84	1649	$\varepsilon\beta^+, 2\varepsilon$	$PbMoO_4$, $LiMoO_4$, $CaMoO_4$
^{100}Mo	9.63	3034	$2\beta^-$	$PbMoO_4$, $LiMoO_4$, $CaMoO_4$
^{84}Sr	0.56	1786.8	$\varepsilon\beta^+$	$SrCl_2$, $SrI_2(Eu)$

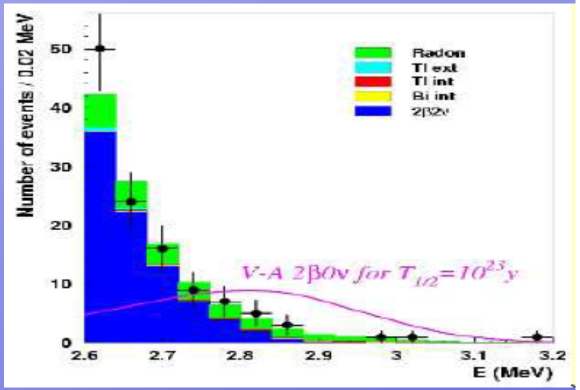
Good Energy Resolution ?



from S. Elliott and P. Vogel

Very helpful to separate $0\nu\beta\beta$ from $2\nu\beta\beta$ and to separate $0\nu\beta\beta$ signal from other γ lines

However, relatively modest energy resolution can be acceptable for most decay modes and, in particular:



- **to separate $0\nu\beta\beta$ from $2\nu\beta\beta$ when fitting the endpoint shape (procedure adopted e.g. in NEMO-3)**
- **to separate $0\nu\beta\beta$ signal from other γ lines in case of low-level background:**

choosing a high Q-value isotope
using an ultra-low background detector and set-up

a distinctive peak can be pointed out from a low-level continuum background

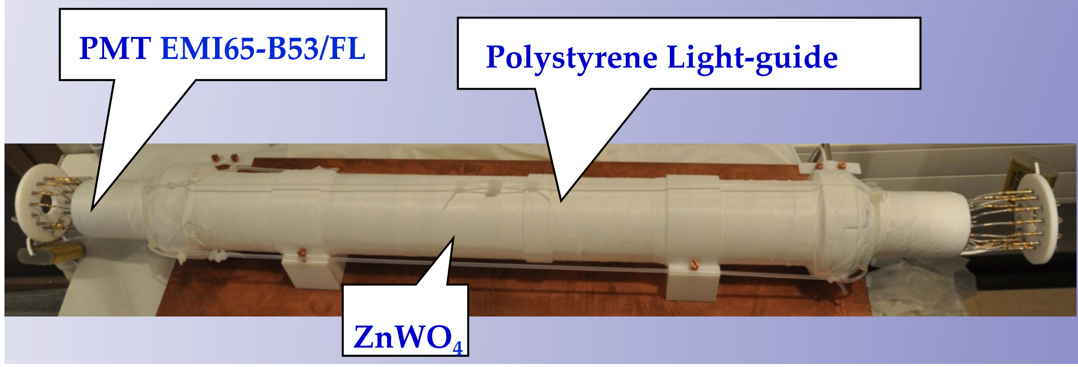
STATUS and PERSPECTIVES on:

Talk by Cerulli

- ✓ $ZnWO_4$ crystal scintillators to search for 2β in Zn and W isotopes
- ✓ $^{106}CdWO_4$ crystal scintillator to search for 2β in ^{106}Cd ← Talk by Tretyak
- ✓ $^{116}CdWO_4$ crystal scintillators to search for 2β in ^{116}Cd ← Talk by Poda

→ Towards larger source=detectors and enrichments to increase sensitivity

\+ many other interesting developments by DAMA-Kiev and coll. in DAMA set-ups



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^-

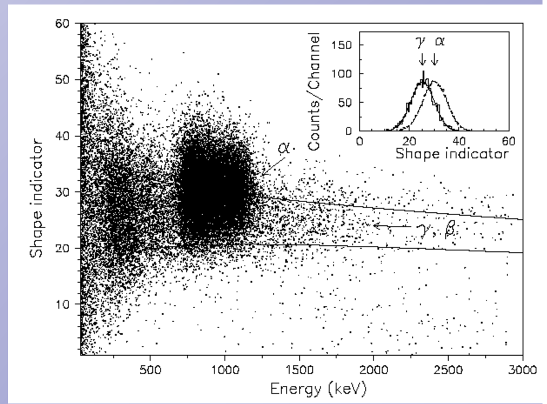
CeF_3 — NIMA 498 (2003) 352
$CeCl_3$ — NPA 824 (2009) 101; JPG:NPP 38 (2011) 015103

CeF_3 crystal scintillators

(2 x 2 x 2) cm^3, mass 49.3 g, produced in China; E_{thr} = 20 keV; σ/E=22% at 122 keV

(14 x 2 x 2) cm^3 , 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) cm^3 , mass 74.5 g, produced in China; E_{thr} ~ 20 keV; σ/E=19% at 122 keV

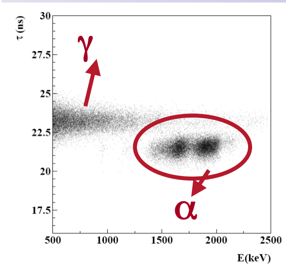


Radioactive contaminations in the CeF_3 crystal scintillator obtained by fitting the experimental spectrum by the background model. The derived in this way activity of ^{232}Th 55(30) mBq/kg is consistent with the value of 37(16) mBq/kg obtained by the pulse-shape analysis of the data

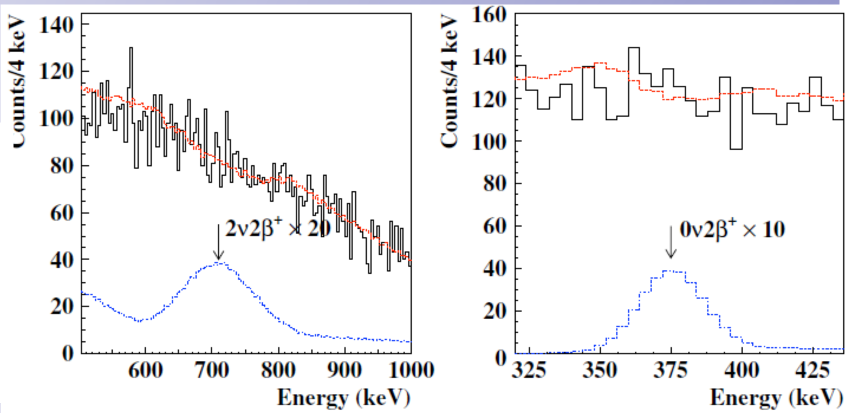
Chain	Source	Activity (mBq/kg)
^{232}Th	^{232}Th	55(30)
	^{228}Ra	890(270)
	^{228}Th	1010(10)
^{238}U	^{238}U	⩽70
	^{234}U	⩽60
	^{230}Th	⩽60
	^{226}Ra	⩽60
	^{210}Pb	⩽280
^{235}U	^{235}U	⩽40
	^{231}Pa	⩽50
	^{227}Ac	⩽20
	^{40}K	⩽330
	^{138}La	⩽60
	^{176}Lu	⩽20
	^{147}Sm	⩽80

$CeCl_3$ crystal scintillators

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



2ν2β$^+$ e 0ν2β$^+$ in ^{136}Ce



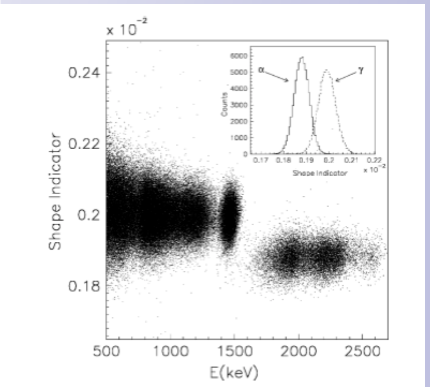
Transition	Decay channel	Decay mode	Exp. $T_{1/2}$ limit (yr) Present work	Previous results
^{136}Ce → ^{136}Ba	$2\beta^+$	0ν	> 0.7(2.5) × 10^{17}	> (6.9) × 10^{17} [9]
		2ν	> 0.9(1.8) × 10^{16}	> 1.8 × 10^{16} [10]
	$\varepsilon\beta^+$	0ν	> 0.9(2.5) × 10^{17}	> 3.8 × 10^{16} [10]
		2ν	> 2.4(5.4) ×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

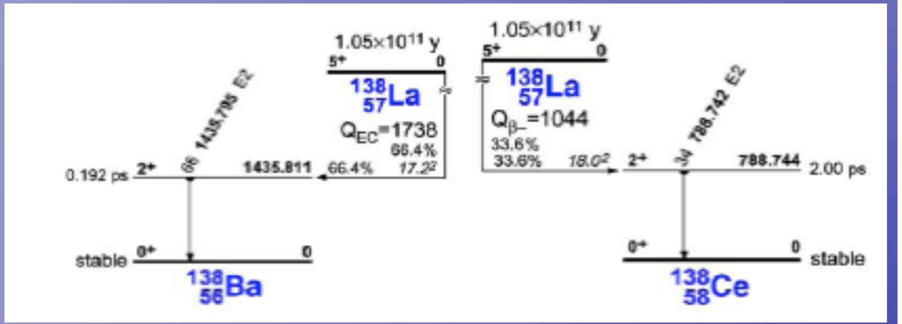
New limits and new perspectives for future measurements: work on radiopurity, enlarge mass, apply enrichement, improve response features, enlarge running time..

$LaCl_3(Ce)$

NIMA555(2005)270
Ukr. J. of Phys 51 (2006) 1037

- ✓ Limitation of La compounds in some low background applications when the energy region of interest approaches the one of the ^{138}La contribution.
- ✓ Presence of daughters from broken ^{235}U.
- ✓ The chemistry of Lanthanides and Actinides is similar, so the U and Th content in Lanthanide minerals can be high. Moreover, even if the decay chains would originally be in equilibrium, U and Th would be removed during the Lanthanide purification, but this procedure is not so efficient for some of the daughters; thus, a trace of U and Th can be kept in form of ^{227}Th, ^{223}Ra and daughters with a similar activity





^{138}La activity in the LaCl3(Ce): (21.1 ± 1.4) Bq

… and $LaBr_3(Ce)$?

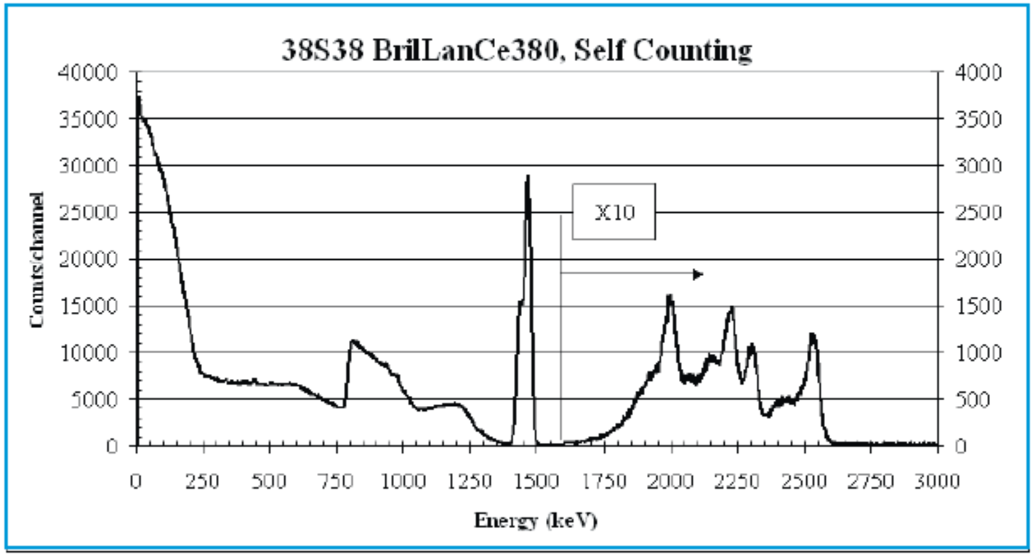


Figure 11
Self-counting background count to 3000 keV

Saint Gobain

Very interesting features for a wide set of applications, but some intrinsic limitations in low background physics, may they be somehow overcome?

Additional interests to develop crystal scintillators with Ce

NPA824(2009)101

- $^{136}Ce \rightarrow ^{136}Ba$ 0ν ECEC resonance:

Isotope	abundance	atomic mass difference	energy (keV) of candidate excited State	energy of K shell of daughter nuclide (keV)	L1	L2	L3
136Ce	0.20	2418.9+/-13	2399.87 (1+,2+) 2392,1 (1+,2+)	37.44	5.98	5.62	5.25

- Excited transitions; expectations from theory:

$\beta^+\beta^+$	g.s.	0ν	(2.6-2.7) x 10^{29}	QRPA	Aun98
$EC\beta^+$	g.s.	2ν	6.0 x 10^{23}	SU(4)	Rum98
ECEC	g.s.	2ν	(3.2-5.1) x 10^{21} 9.6 x 10^{21}	SSDH SU(4)	Civ98 Rum98

New limits and new perspectives for future measurements and detection of some channels reachable: work on radiopurity, enlarge mass, fragmented set-up, improve response features, enlarge running time, apply enrichement, ..

Large space to reach competing results

A further interesting example for future developments: $SrI_2(Eu)$

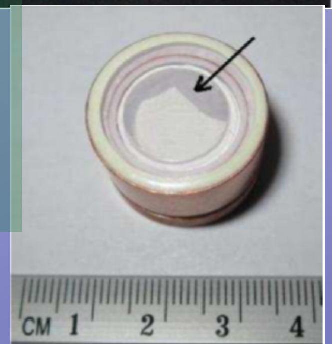
- High light output (>100000 g/MeV)
- Good energy resolution (~3% at 662 keV)
- Absence of natural long-living radioactive isotopes.

NIMA670(2012) 10

Properties of $SrI_2(Eu)$ crystal scintillators.

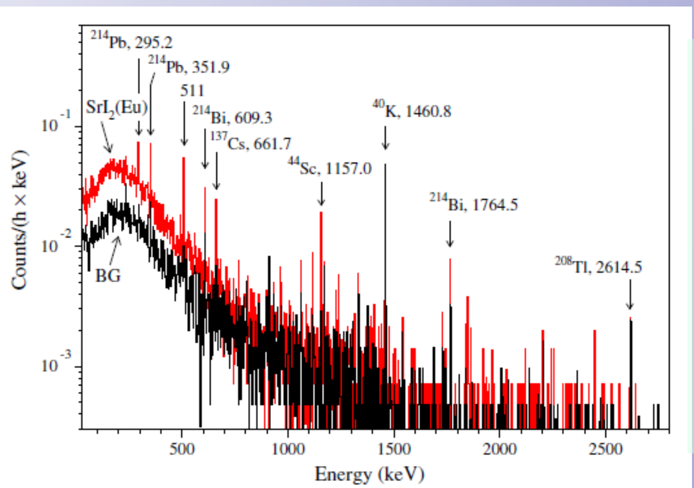
Property	Value
Density (g/cm^3)	4.5–4.6
Melting point (°C)	515
Structural type	Orthorhombic
Index of refraction	1.85
Wavelength of emission maximum (nm)	429–436
Light yield (photons/MeV)	(68–120) $\times 10^3$
Energy resolution (FWHM, %) for 662 keV γ of ^{137}Cs	2.6–3.7
Scintillation decay time (μs)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

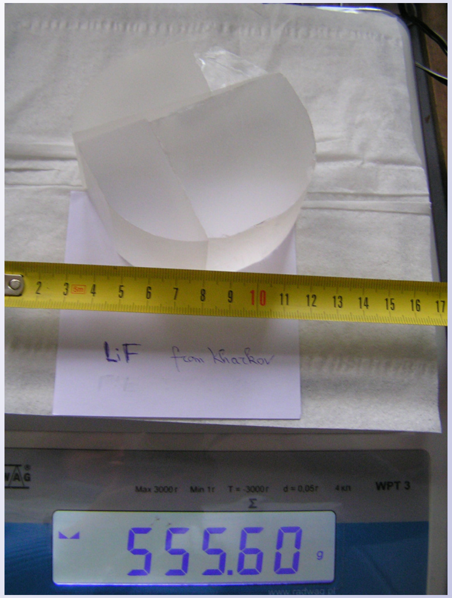


Measurement in the Ge facility at LNGS

Measured during 706 h with ULB-HPGe to investigate bckg and set 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 larger crystal mass, longer meas. time and source=detector → appreciable sensitivity expected

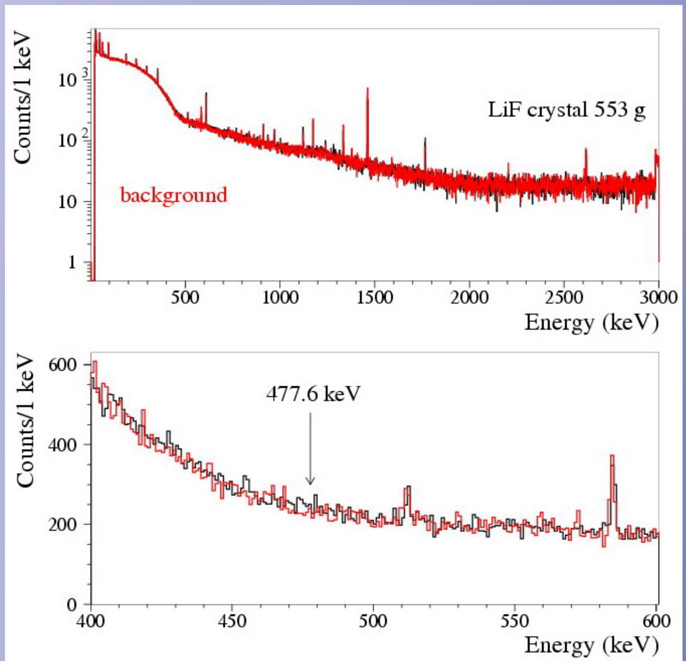


U/Th contamination < ~0.01 Bq/kg

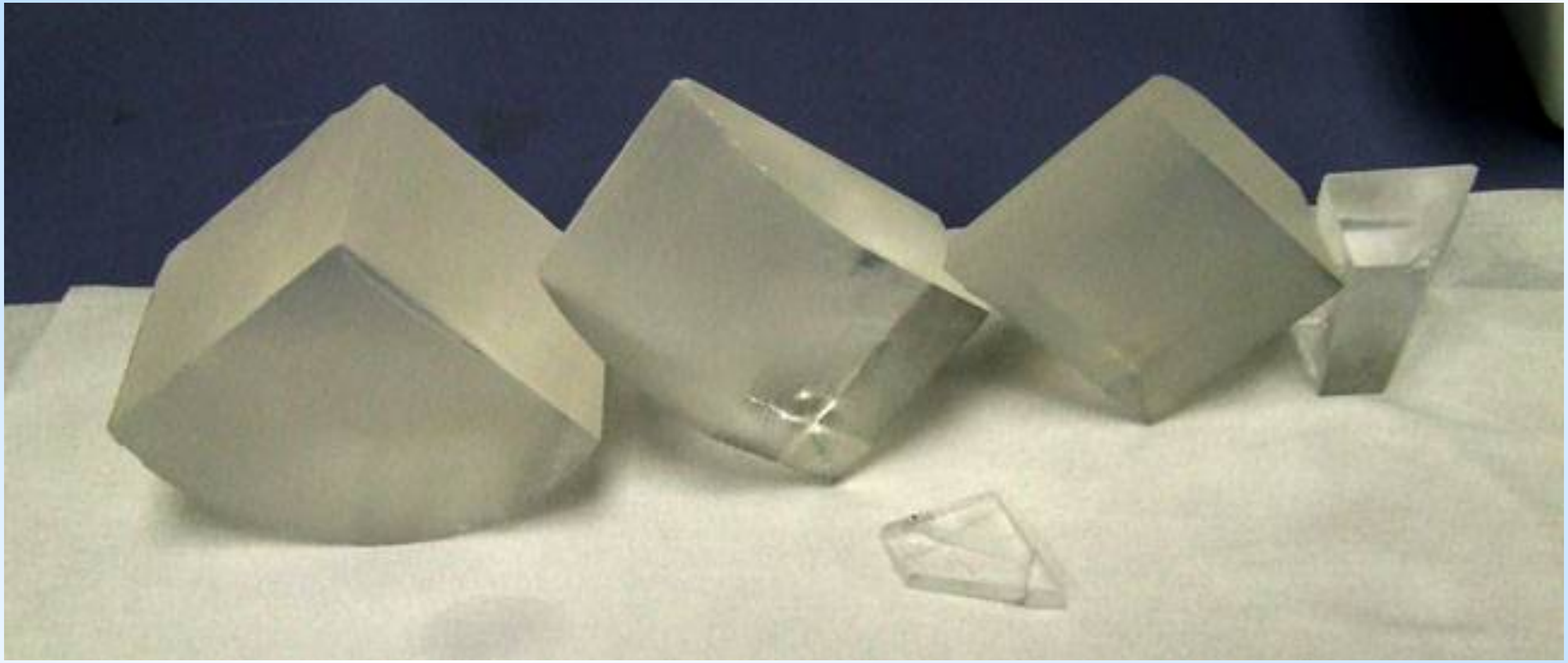
LiF(W) for ^{7}Li solar axions

PLB711(2012)41

- a LiF(W) crystal (553 g) in measurement in GEBER (244 cm3) in 4044 h
- The crystal was produced by the Czochralski method.
- To improve its scintillation properties, it is doped by W.
- Comparison of the measured LiF(W) spectrum with the HPGe background shows that — in contrast with the LiF powders — the LiF(W) crystal is very pure: the energy distribution measured with the sample practically coincides with the bckgr
- Only limits for radioactive pollutions were derived



LiF crystals

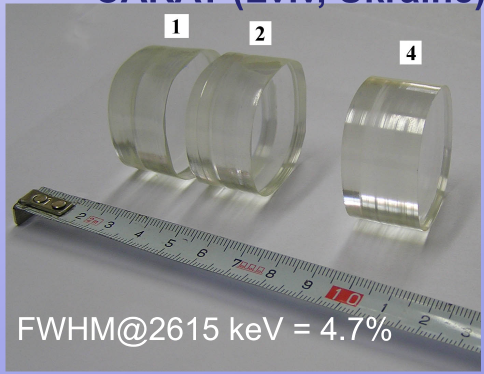


Encourage possible developments of doped LiF scintillators with suitable light response for some applications

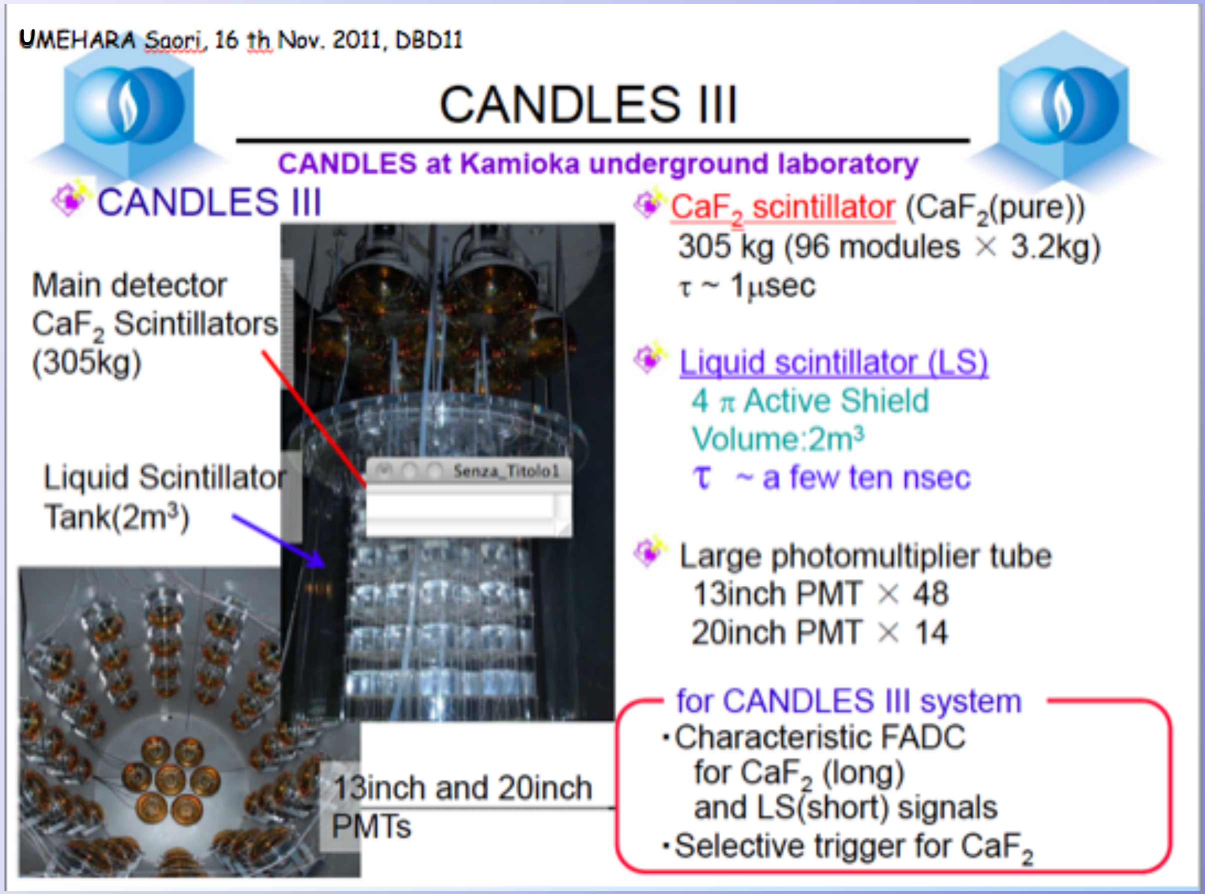
Among the many existing efforts

✓ $CaMoO_4$ crystals at Y2L → next speaker

$CaMoO_4$ ∅40×20 mm, CARAT (Lviv, Ukraine)

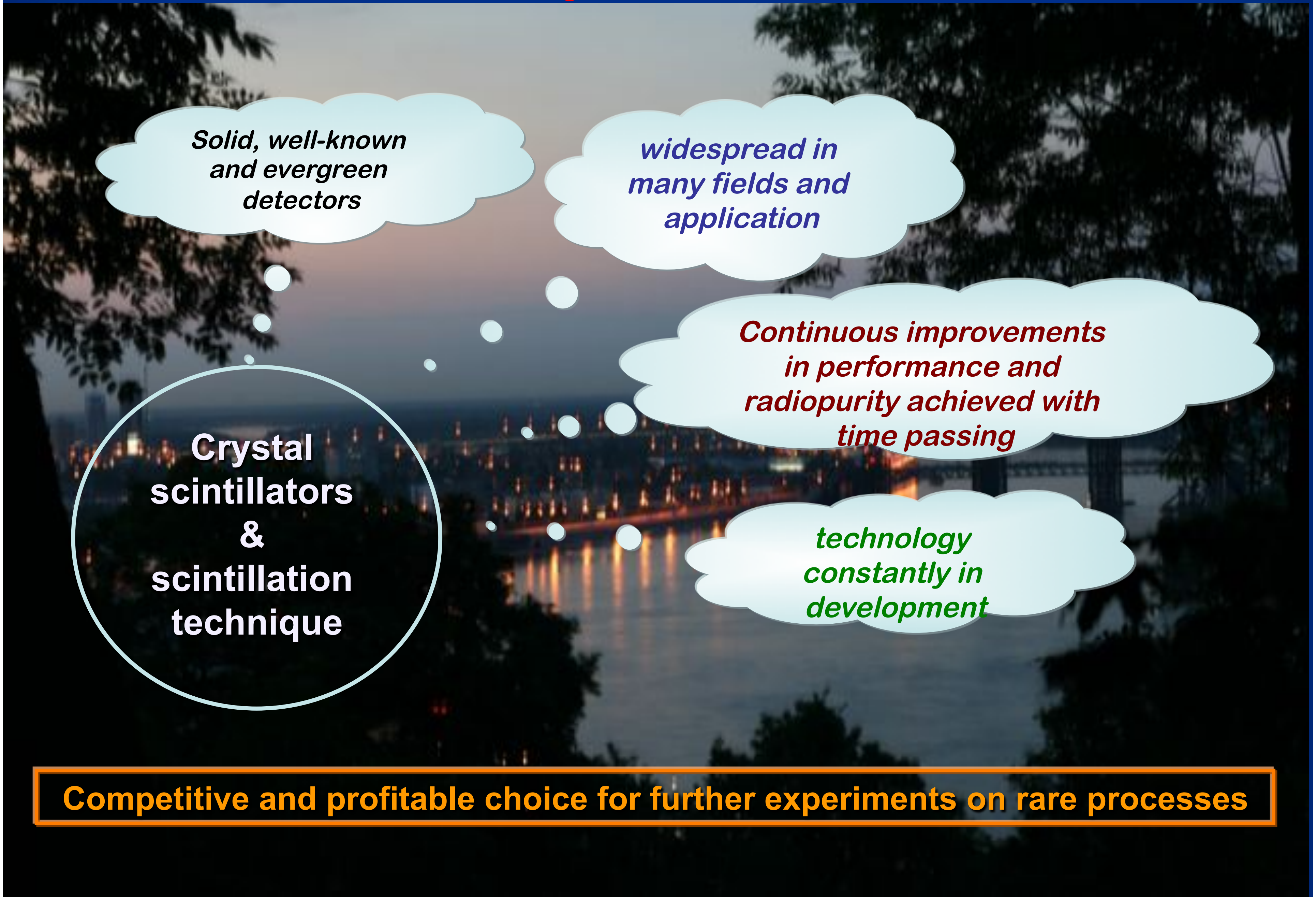


✓



✓ … and at cryogenic T → dedicated talks at this workshop

...Conclusion...



Solid, well-known and evergreen detectors

widespread in many fields and application

Continuous improvements in performance and radiopurity achieved with time passing

technology constantly in development

Crystal scintillators & scintillation technique

Competitive and profitable choice for further experiments on rare processes