Development of tungstates, molybdates and chalcogenides scintillation crystals for HEP and REP

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REQUIREMENTS TO SCINTILLATORS

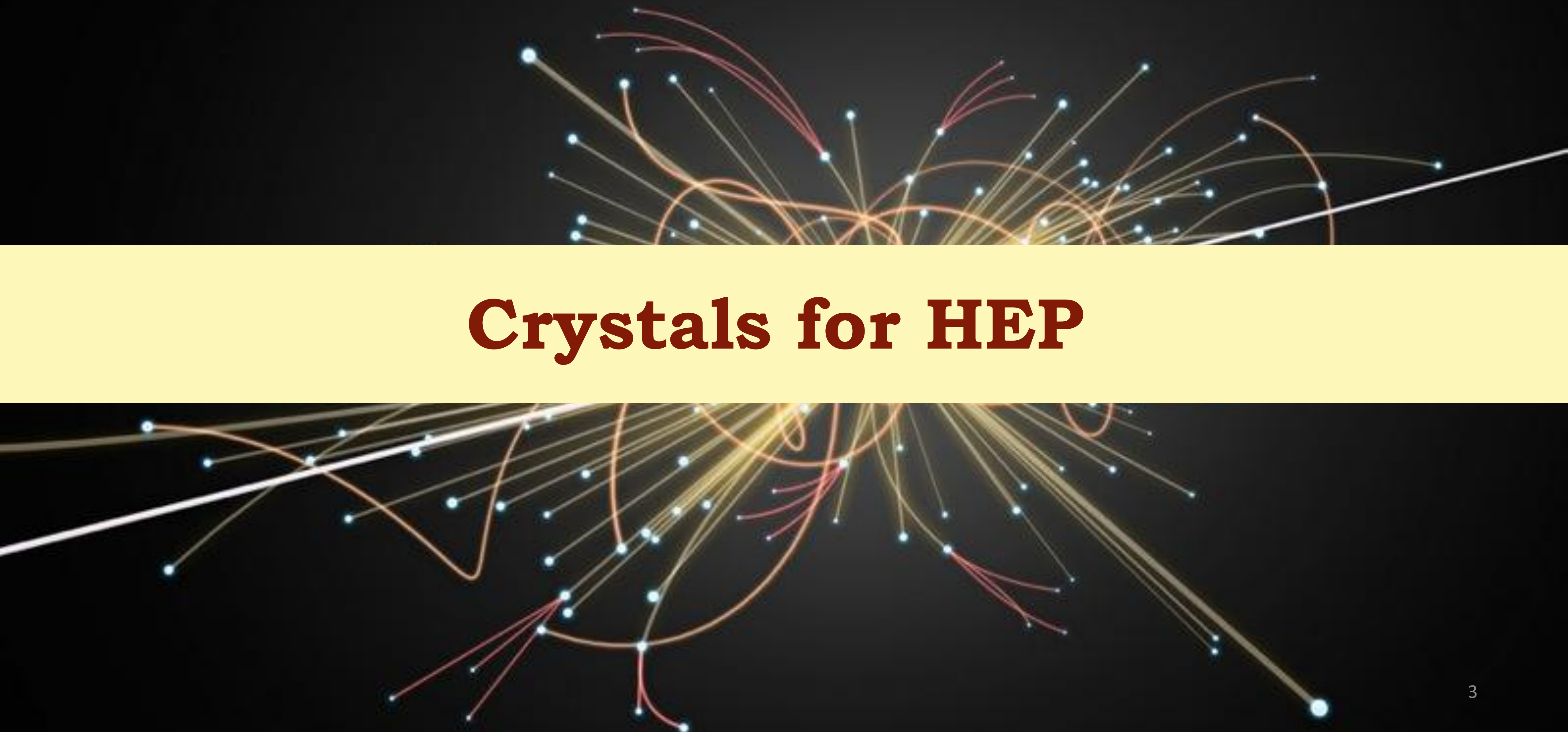
for High Energy Physics (HEP)
EM calorimetry

- high radiation hardness
- high Z_{eff}, density (short radiation length)
- high light output
- short decay time
- high optical quality
- possibility of tonnage production (hundreds of tons)
- low cost

for Rare Event Physics (REP)
cryogenic scintillating bolometer

- extremely low level of radioactive contamination
- presence of specific nuclei (2b decay) (for example $^{64,70}Zn$, $^{180,186}W$, $^{92,98,100}Mo$, ^{82}Se, ^{130}Te)
- variety of elements in scintillation targets (Dark matter)
- high light output at milli-Kelvin temperature
- high optical quality
- high crystalline structure perfection

Crystals for HEP

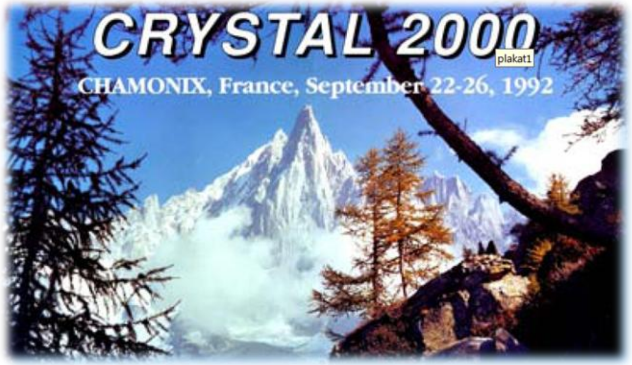


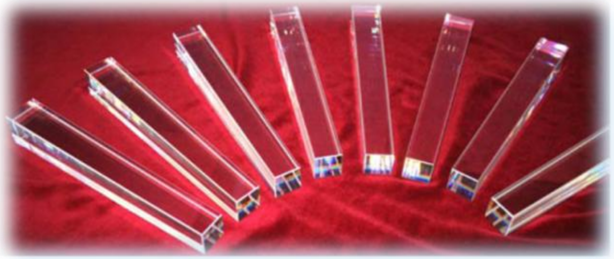
PWO for CMS

Scintillators used in HEP calorimeters

	NaI(Tl)		BaF_2		CsI(Tl)		CeF_3		BGO $Bi_4Ge_3O_{12}$		PWO $PbWO_4$	
X_0, sm	2,59	☹	2,03	☹	1,86	☺	1,66	☺	1,12	☺	0,92	☺
ρ, g/sm³	3,67	☹	4,89	☹	4,53	☹	6,16	☺	7,13	☺	8,2	☺
τ, ns	230	☹	0,6 620	☺ ☹	1050	☹	30	☺	340	😐	15	☺
λ, nm	415	☺	230 310	☺ 😐	550	☺	310 340	😐 😐	480	☺	420	😐
LY, %NaI	100	☺	5 16	☹ ☹	85	☺	5	😐	10	☺	0,5	☹
Experiment	Crystal Ball		L , GEM, TAPS		Belle, BaBar BesIII		L3P		L3, Belle		LHC	

P.Lecoq, et al «Inorganic Scintillators for Detector Systems», Springer Cham, 2017, 408 P., DOI 10.1007/978-3-319-45522-8

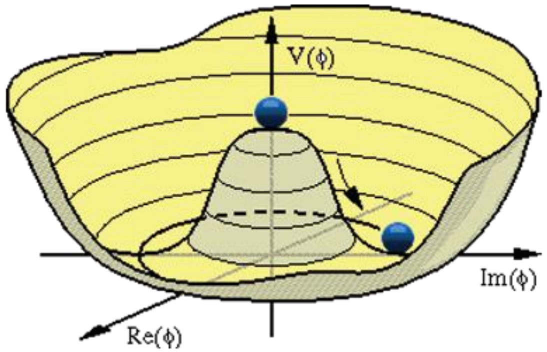


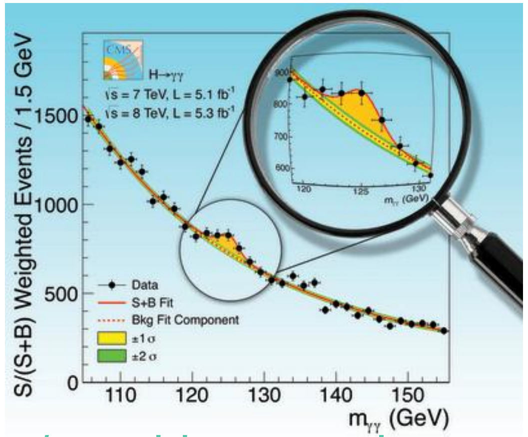


2012 - the Higgs boson has been found

Scientists from the European Center for Nuclear Research (CERN) announced that they have detected a signal of a particle with a mass equal to 125 GeV, that a new subatomic particle corresponding to the parameters of the Higgs boson has been found.

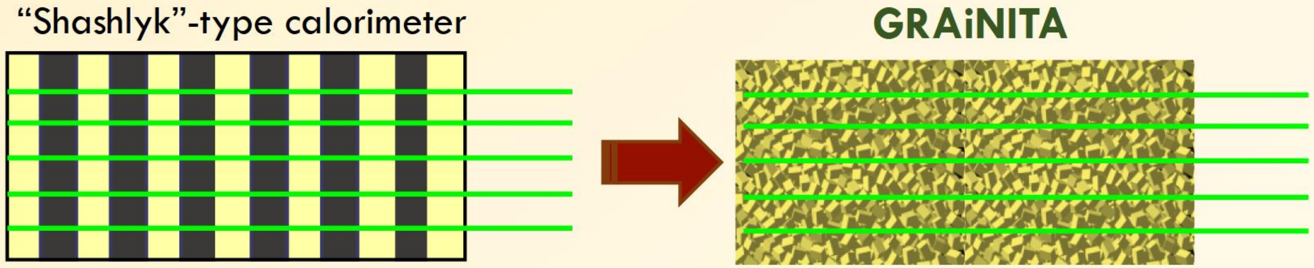
CMS Higgs Physics Results





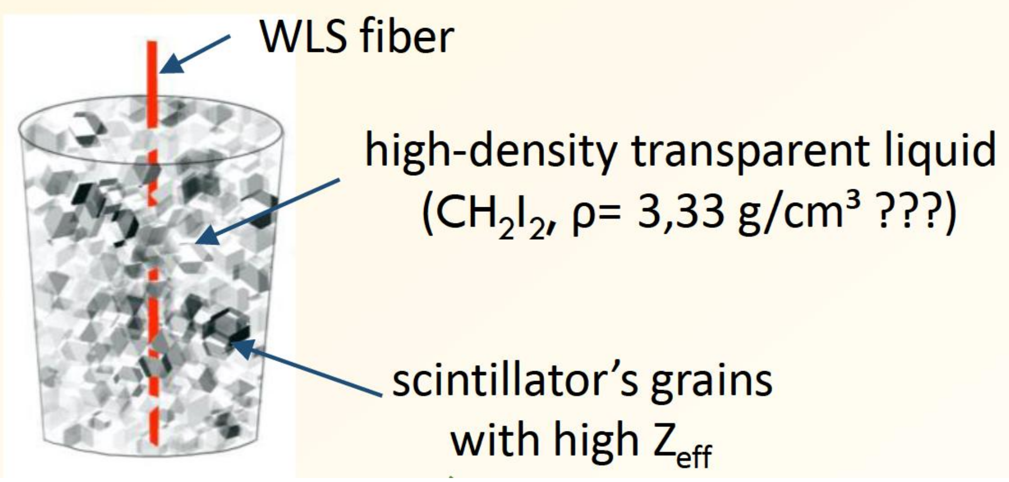
https://cms.cern/physics/cms-higgs-results

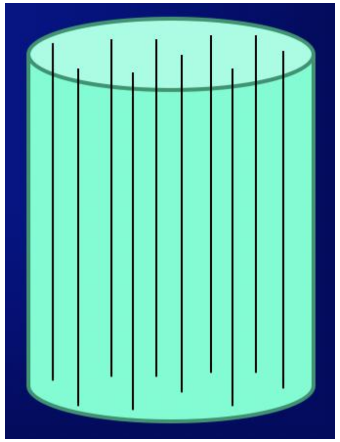
GRAiNITA concept

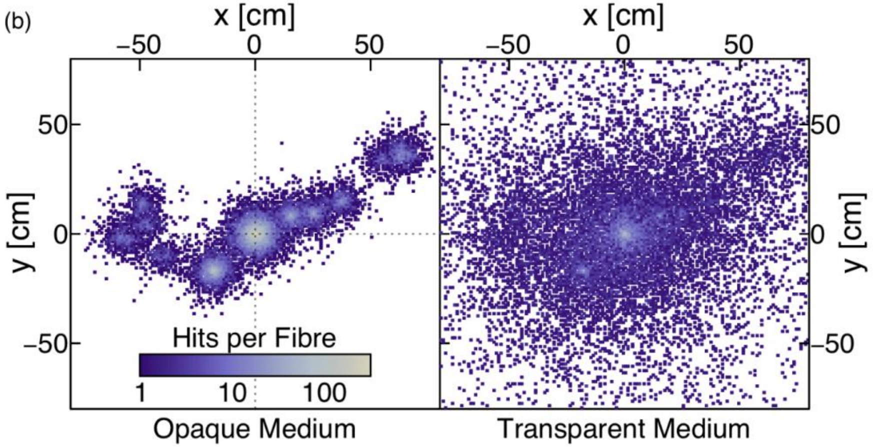


$$\frac{\sigma_E}{E} \sim \frac{10\%-15\%}{\sqrt{E}}$$

$$\frac{\sigma_E}{E} \sim \frac{1\%-2\%}{\sqrt{E}}$$ is expected







$ZnWO_4$

LY= 10 000 ph/MeV

- Z_{eff}=61
- Density 7.85 g/cm³
- Index n=2.1-2.3
- τ = 20 μs
- λ_{max}^{em} = 490 nm

$CaWO_4$

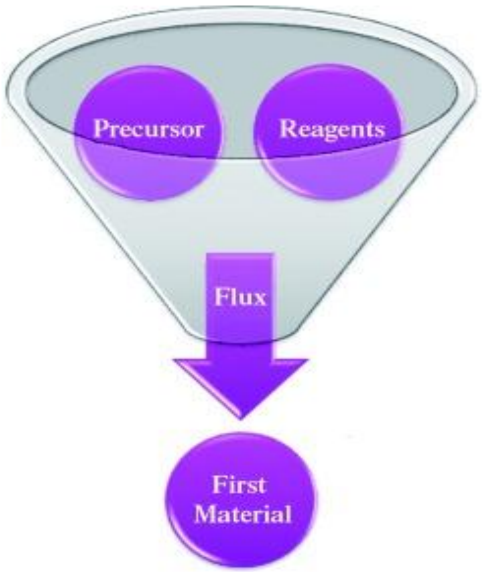
LY= 14 000 ph/MeV

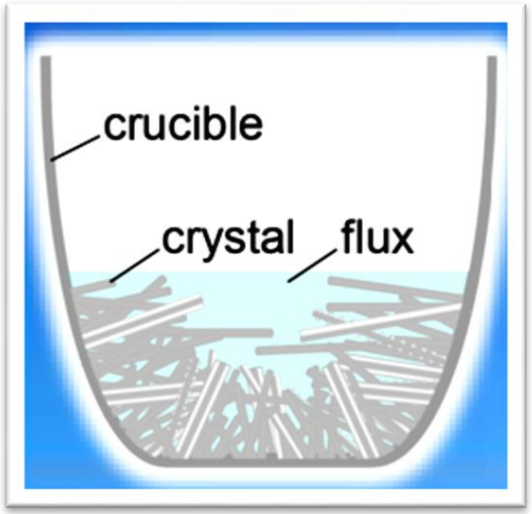
- Z_{eff}=62
- Density 6. 01 g/cm³
- Index n= 1.9
- τ = 12 μs
- λ_{max}^{em} = 420 nm

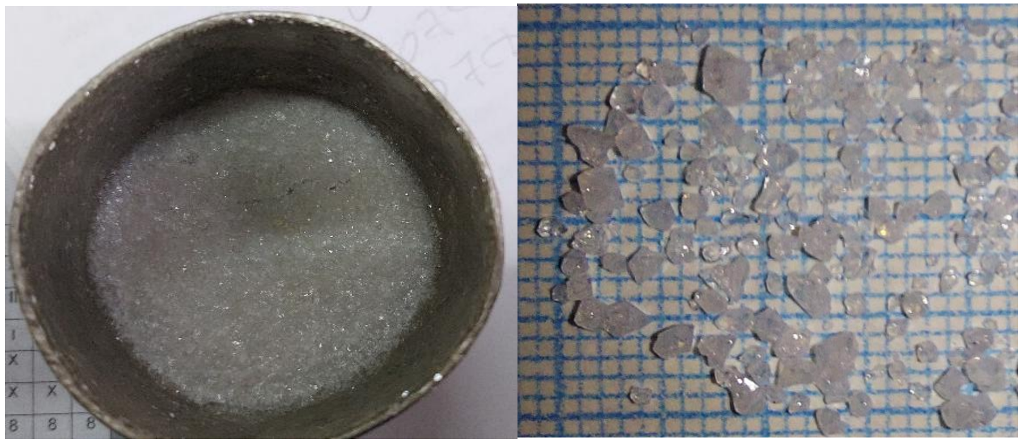
Inspired by LiquidO technique for neutrino detector (A. Cabrera et al. LiquidOCommun Phys 4, 273 (2021))

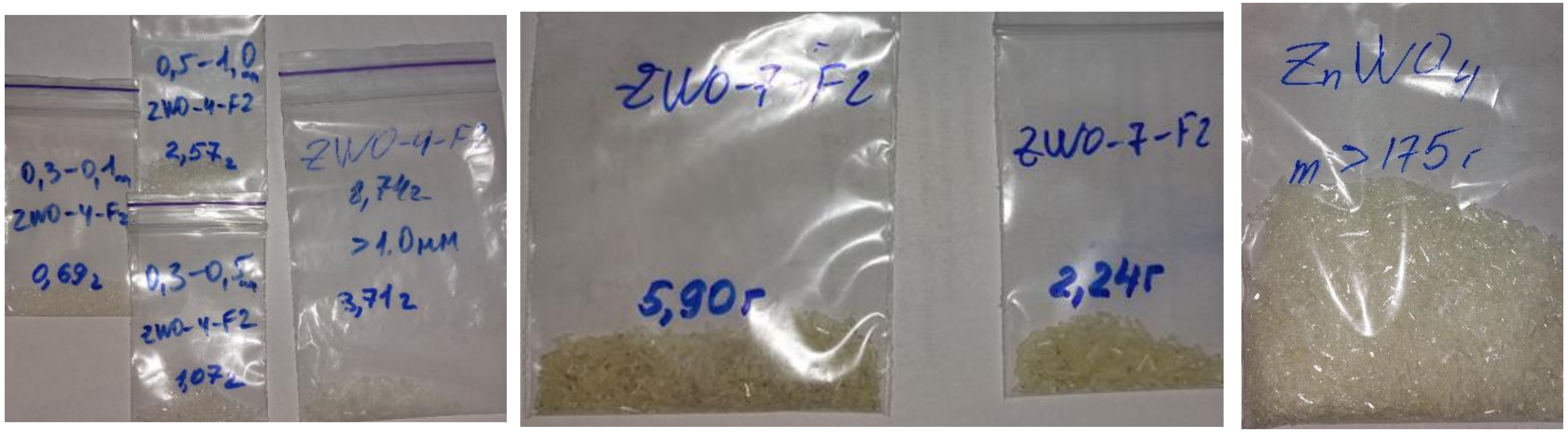
S.Barsuk, et al, GRAiNITA – a new generation calorimeter, Réunion GDR DI2I, 10-12 julliet 2023, Subatech, Nantes, France, https://indico.in2p3.fr/event/29808/contributions/126587/

The flux method





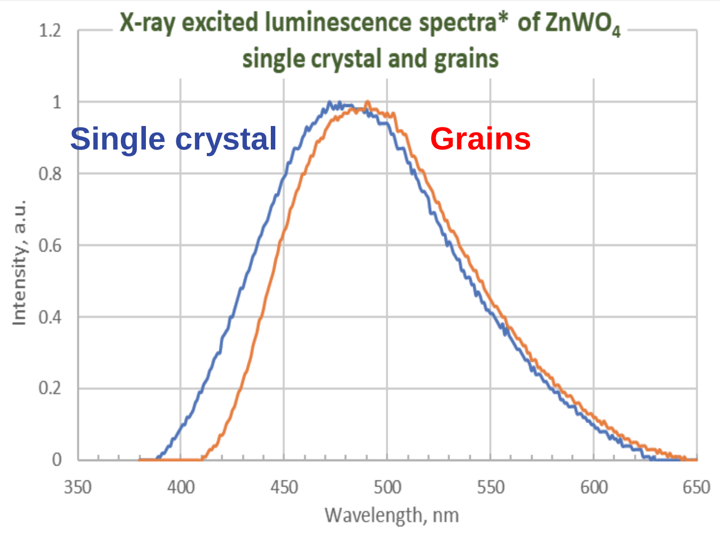


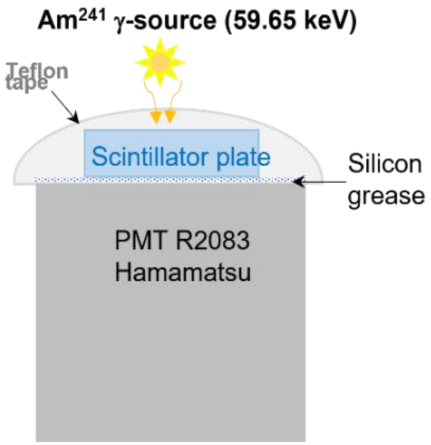


$ZnWO_4$ crystal grains obtained via the flux method:
a) $NaCl + ZnWO_4$, b) $Na_2WO_4 + ZnO + WO_3$, c) $Na_2WO_4 + ZnWO_4$

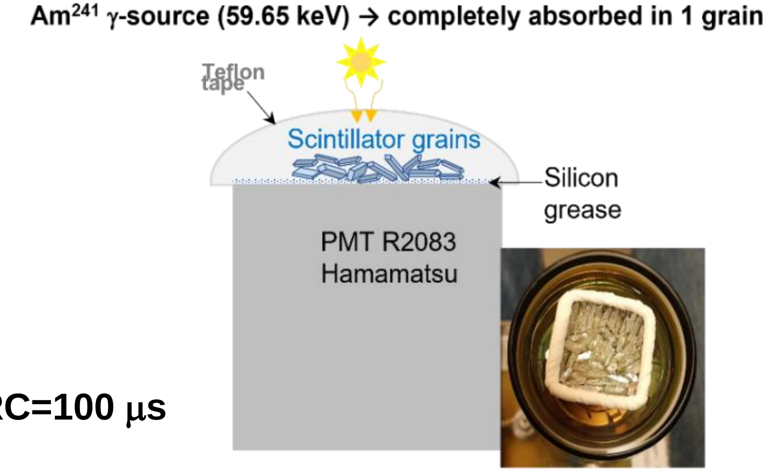
Scintillation characteristics of $ZnWO_4$ grains

$ZnWO_4$ crystal grains obtained via the flux method



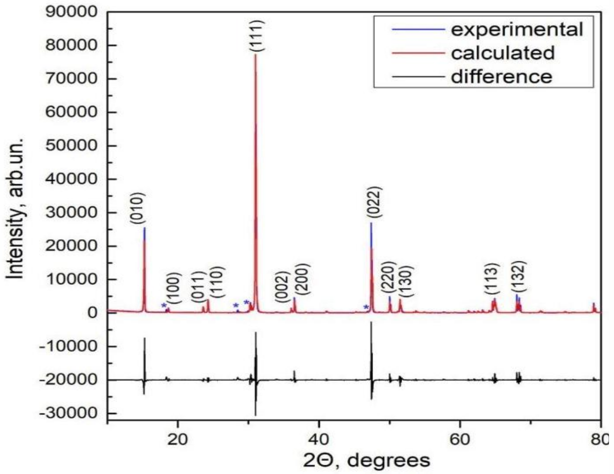


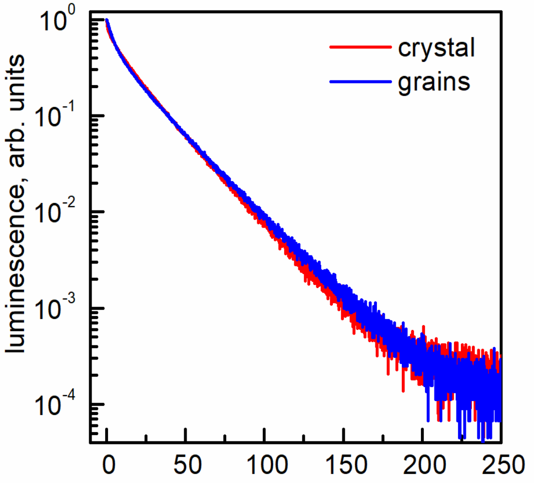
RC=100 μs

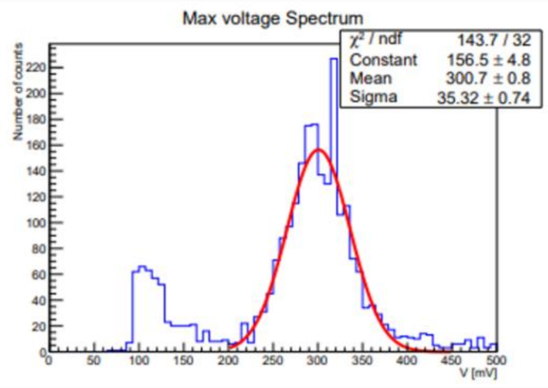


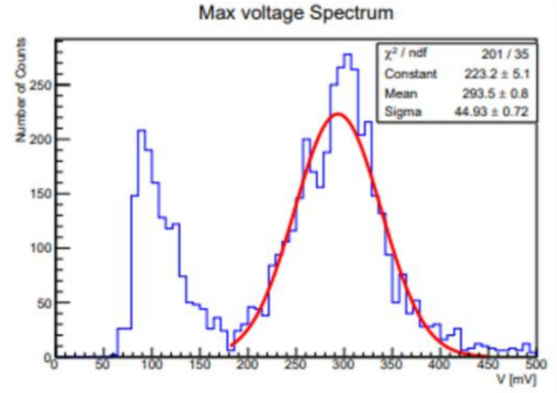
$ZnWO_4$ single crystal plate 20x20x1 mm^3

$ZnWO_4$ grains "mosaic" ~20x20x1 mm^3



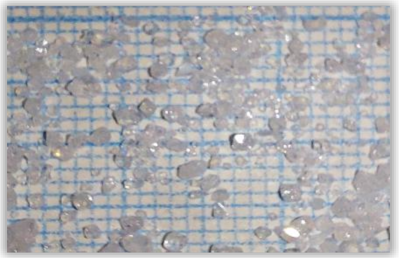




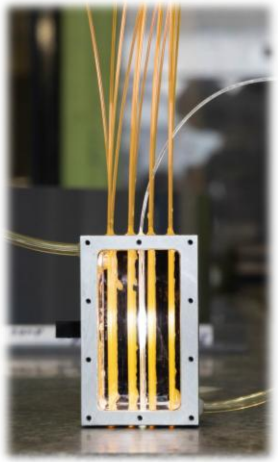


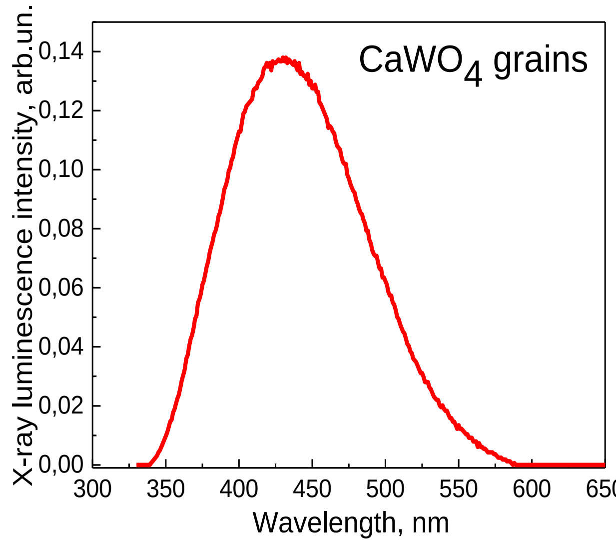
- Good reproducibility of values measured for grains from $ZnWO_4$ crystal grains batches → stable technology for grains production
- The batches of grains show a much smaller variance in the amplitude of the Am^{241} peak at 59.65 keV → the better homogeneity in the light yield

$CaWO_4$ scintillation single crystal grains for use in the GRAiNITA project detector

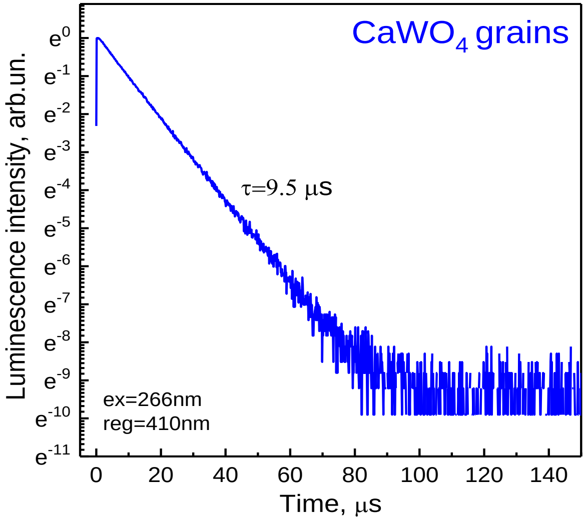


The small-size prototype





X-ray luminescence of $CaWO_4$ grains



Photoluminescence decay curves of $CaWO_4$ grains

Charge $CaWO_4$ | **Na_2WO_4**

↓

Mixing
Planetary mill

↓

Growing granule $CaWO_4$ using by flux method:
Heated – from 20°C to 1200°C.
Held - 1200°C. Cooled – from 1200°C to 600°C.
Cooling resistance furnace to room temperature.

↓

Separation of grains with distilled water at 100°C .
Separation of grains with distilled water at 60°C in ultrasonic cleaner.

↓

Annealing of crystals.
T=800°C.

	$CaWO_4$ published crystallographic data	$CaWO_4$ grains crystallographic data
Crystal system	tetragonal	tetragonal
a, Å	5.24294	5.24086
c, Å	11.373	11.37239
Cell volume, Å^3	312.63	312.36

Troll characterization at CERN

Crystals for REP

$ZnWO_4$ is one of the best candidate as a detector for cryogenic 2β decay and Dark Matter experiments

Large volume $ZnWO_4$ crystal

Activity (mBq/kg)

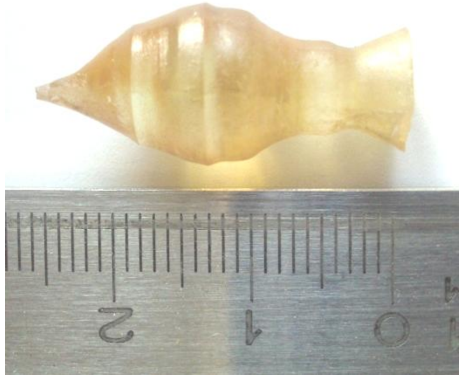
	$ZnWO_4$ (1)	$CaWO_4$ (2)
^{232}Th	0.0015	0.7
^{235}U	≤ 0.003	1.6
^{238}U	0.002	300
Total α	0.18	400
^{40}K	≤ 0.4	
^{65}Zn	0.5	
^{90}Sr-^{90}Y	≤ 0.4	

1) P. Belli, et al., NIM A 626-627 (2011) 31

2)NIMA 538 (2004) 657; APP 23 (2005) 249

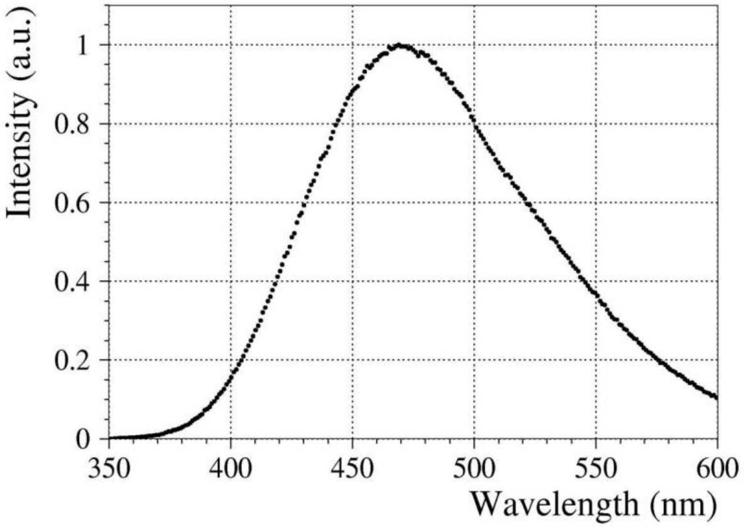
The relative light output of $ZnWO_4$ at 10 K is 110-115 % vs $CaWO_4$

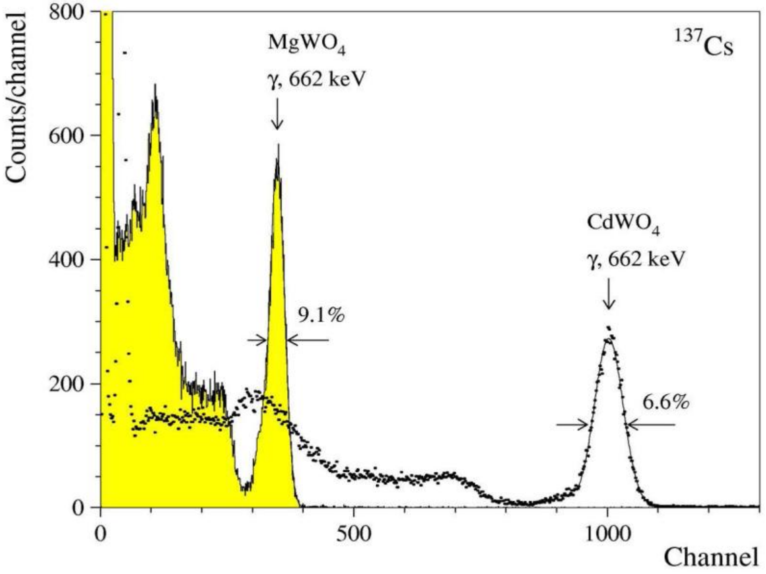
$MgWO_4$ crystals for Dark Matter search



Cryogenic dark matter experiments require a variety of scintillation targets to verify the nature of a detected signal. The main difficulty of crystal growth from the melt is the presence in $MgWO_4$ of a phase transition below the melting point. Magnesium tungstate crystals of a few cm^3 volume were grown for the first time by pulling the seed from the melted flux solution.

XRL and Energy spectra of 662 keV ^{137}Cs γ-rays measured for $MgWO_4$





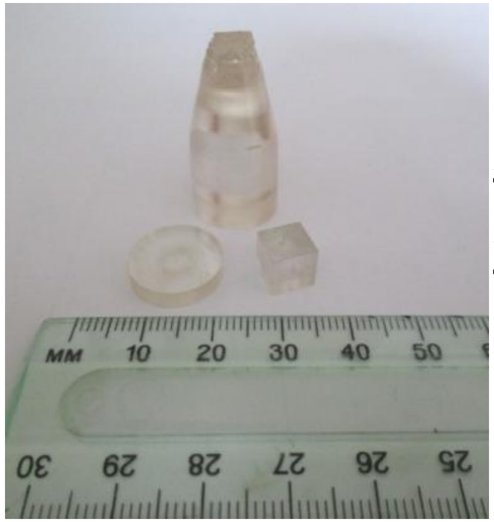
The crystal shows intense luminescence under X-ray excitation. The broad emission band exhibits maximum at 475 nm.

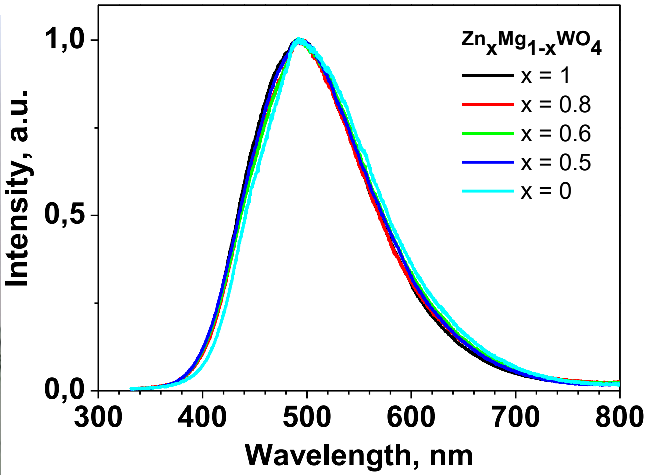
The relative photoelectron yield found to be 35% vs $CdWO_4$ and 27% vs NaI(Tl).

The energy resolution of magnesium tungstate crystal scintillator giving 9.1 % for the 662 keV γ -quanta of ^{137}Cs

$Zn_xMg_{1-x}WO_4$ scintillators

The set of single crystals of $Zn_xMg_{1-x}WO_4$ (x = 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 0.95) were grown by the Czochralski method





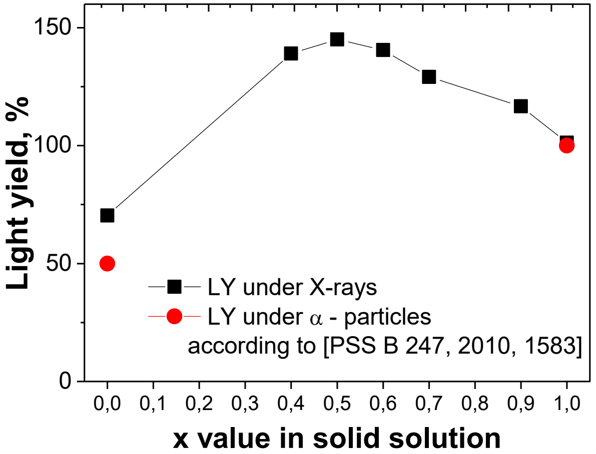
Normalized emission spectra under X-ray excitation, T = 300 K

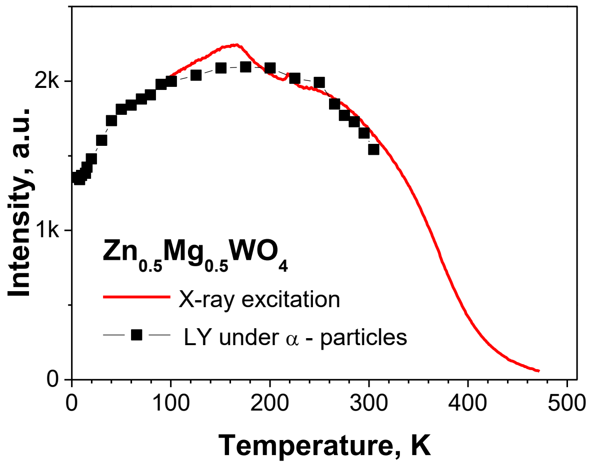
Decay times, E_{ex} = 4.1 eV	
x	τ, mks
1	26
0.9	27
0.8	29
0.7	25
0.6	32
0.5	33
0	39

The single emission band is observed that is due to the emission of exciton, self-trapped on WO_6 complex.

The characteristics of emission center are similar for all of the studied solid solutions.

Light output of $Zn_xMg_{1-x}WO_4$

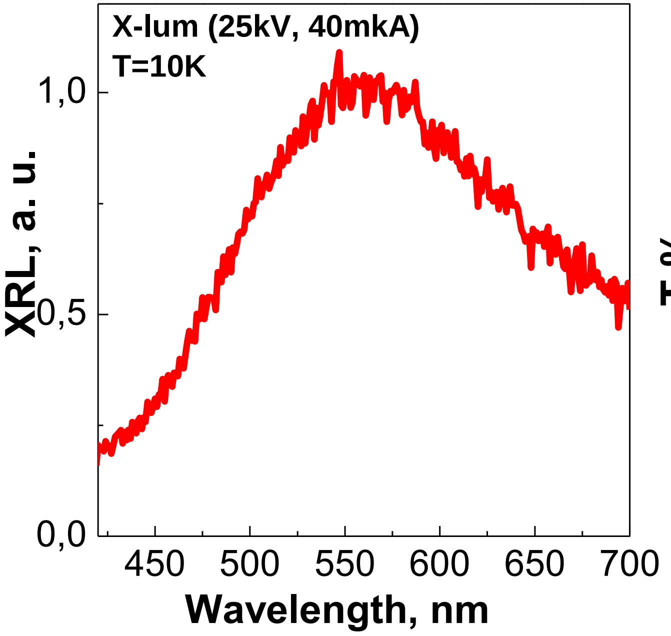


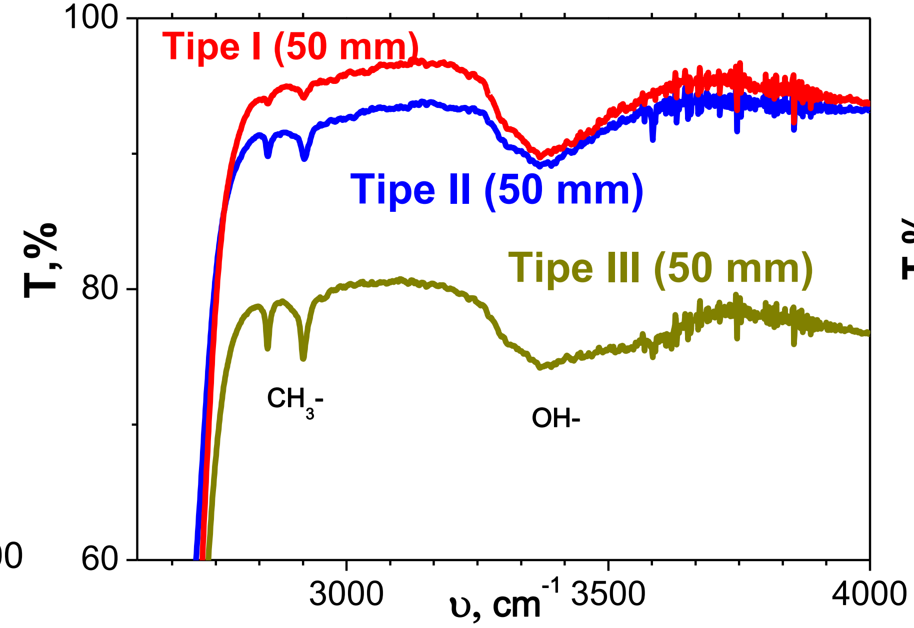


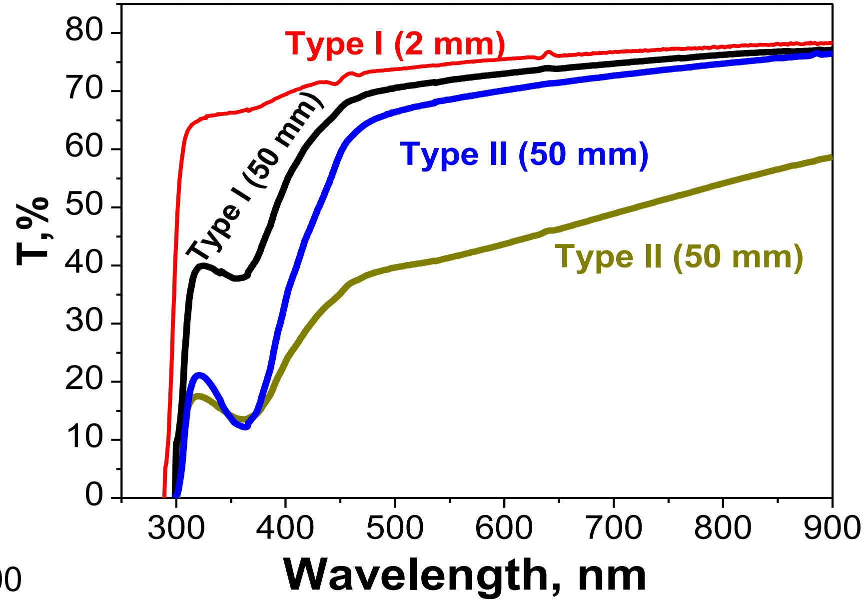
The effect of enhancement of light output is observed for intermediate values of x!

Li_2MoO_4 cryogenic scintillator

Type-I: double crystallization, the dry air, MoO_3 3N and Li_2CO_3 4N.
Type-II: MoO_3 3N and Li_2CO_3 4N, ambient atmosphere.
Type-III: raw materials 2N , ambient atmosphere.

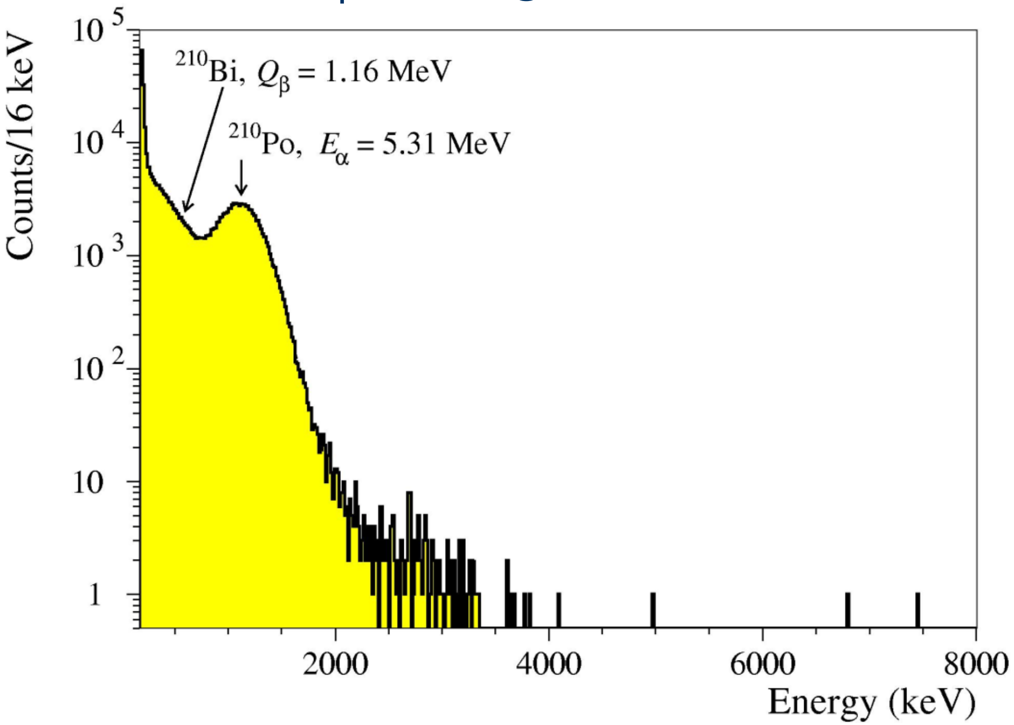






$PbWO_4$ for rare event searches

$PbWO_4$, 182 g, 2.2 h

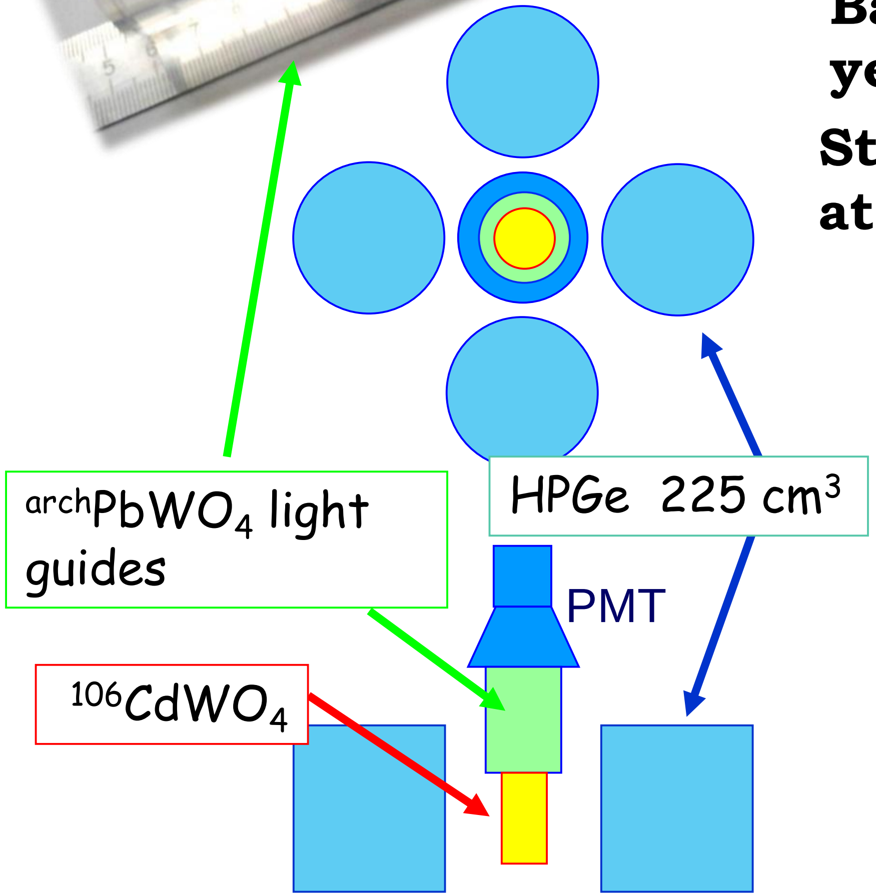


Chain	Nuclide	Activity (mBq/kg)
^{232}Th	^{228}Th	≤13
^{238}U	^{226}Ra	≤10
	^{210}Po	$(53\text{-}79)\times10^3$

F.A.Danevich et al., “Application of $PbWO_4$ crystal scintillators in experiment to search for 2β decay of ^{116}Cd”, NIMA 556(2006)259.

The radioactive background level of these crystals is rather high and is determined by the presence of ^{210}Po. ISMA carried out the work on synthesis of charge and growing of crystals with low radioactive background from the ancient (archaeological) lead.

$^{ARCH}PbWO_4$ for 2β decay of ^{106}Cd with an enriched $^{106}CdWO_4$



4 HPGe, ~ 225 cm³ each,in one cryostat
$^{106}CdWO_4$ in coincidence/anticoincidence with HPGe
Detection efficiency ~ 5 – 7%
Background expected: several events during a year

Starting from 2012 the experiment was made at the Gran Sasso laboratory

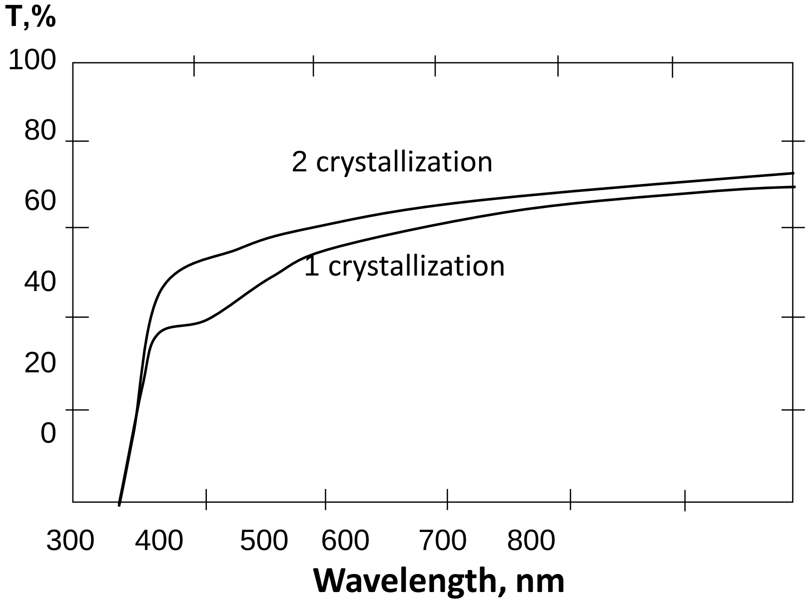
Estimated sensitivity to two neutrino $\varepsilon\beta^+$ and $2\beta^+$ in ^{106}Cd: $T_{1/2} \sim 10^{20} - 10^{21}$ yr

Theory: $2\nu 2K$ $\quad 10^{20} - 5 \cdot 10^{21}$ yr

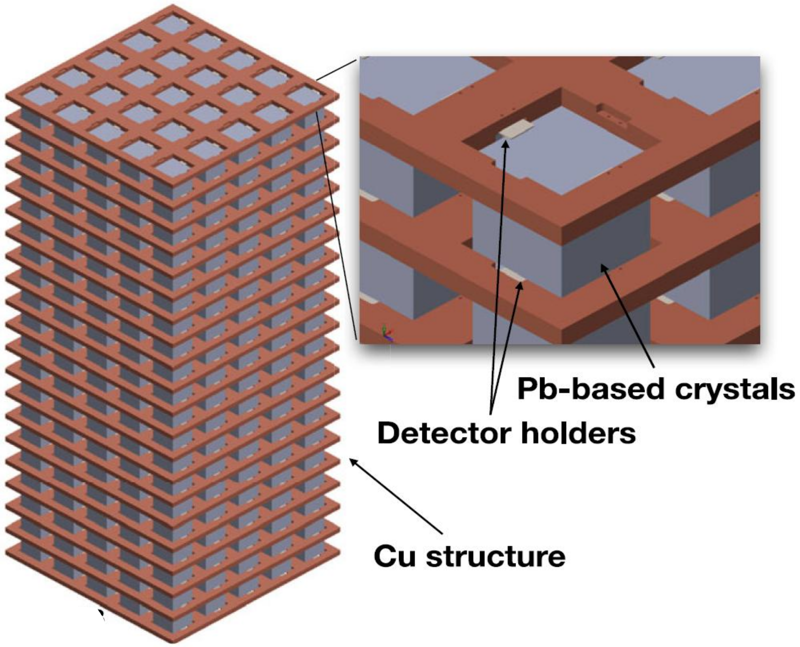
$2\nu\varepsilon\beta^+$ $\quad 8 \cdot 10^{20} - 4 \cdot 10^{22}$ yr

[1] P. Belli et al., NIMA 615 (2010) 301
[2] P. Belli et al., PRC 85 (2012) 044610

Prospects for the use of low-background $^{arch}PbWO_4$ crystals for low background nuclear spectrometry



Coherent Elastic Neutrino-Nucleus Scattering



Cryogenic detector of coherent interaction of neutrinos with $^{arch}PbWO_4$ crystals (2.4 tons, RES-NOVA project) [1]

[1] L.Pattavina et.al., Neutrino observatory based on archaeological lead, Phys. Rev D102 (2020) 063001

The synthesis of $Zn^{82}Se$ charge and crystal growth for CUPID bolometric 0νββ experiment

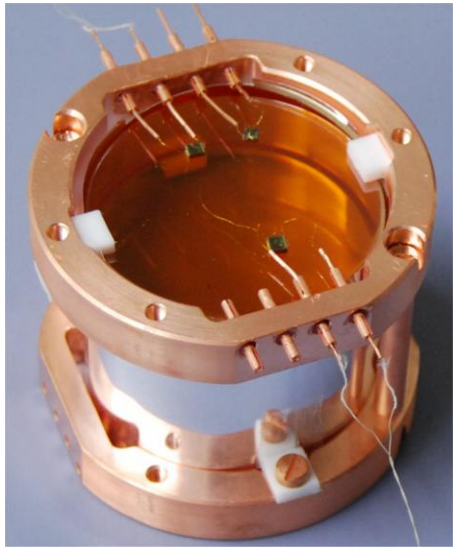
$Zn^{82}Se$ synthesis

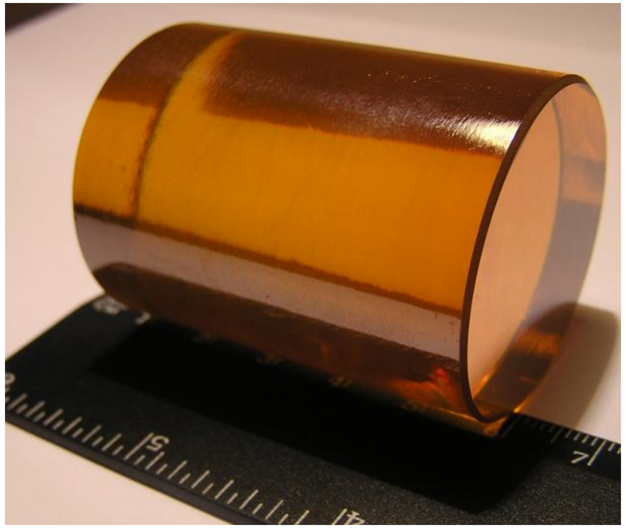
↓

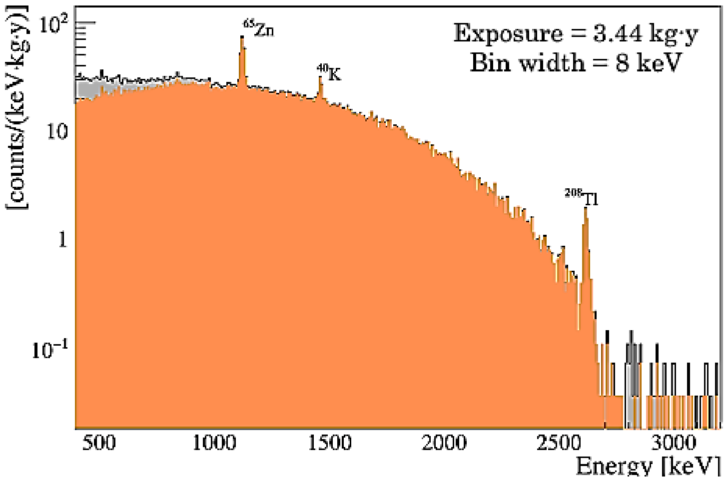
$Zn^{82}Se$ crystal growth

↓

manufacturing of bolometric elements







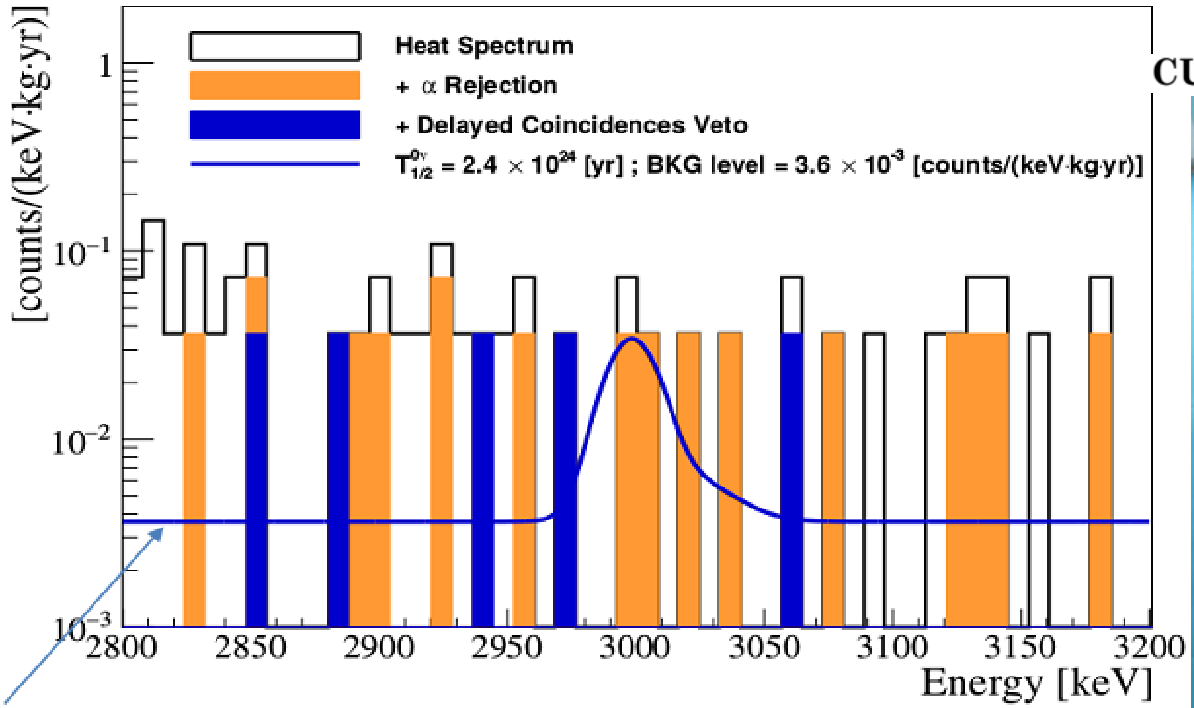
the lowest background level $3.6 \cdot 10^{-3}$ cts/(keV · kg · yr))

CUPID-0 Results: half life and neutrino mass

$$T_{1/2}^{0\nu} > 2.4 \cdot 10^{24} \text{ yr } (90\% \text{ C.I.})$$

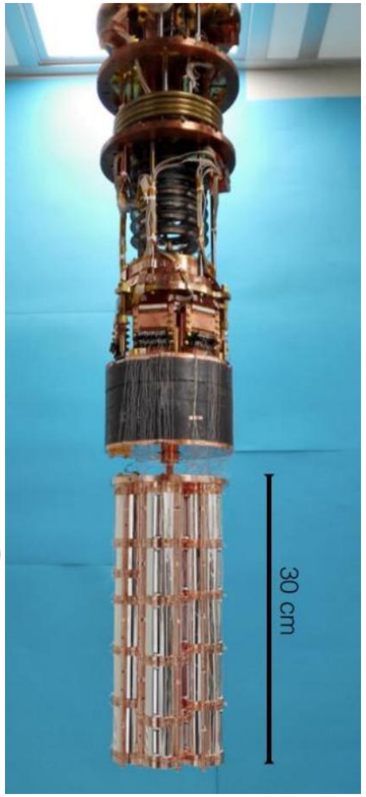
$$m_{\beta\beta} < 376 - 770 \; meV$$

range due to the nuclear matrix element calculations



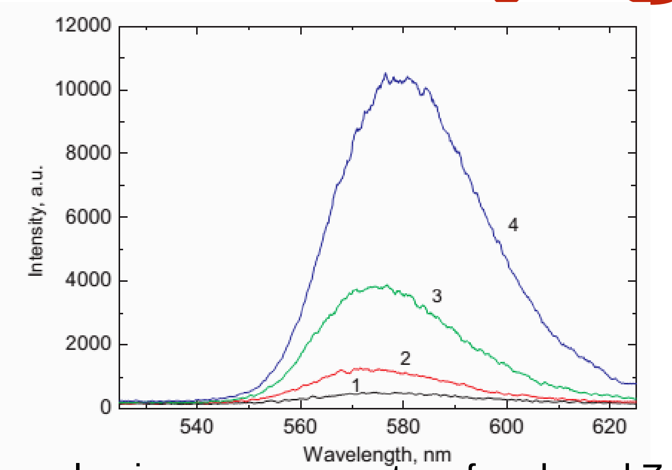
fitted spectrum together with a hypothetical signal corresponding to the 90% C.I. limit

CUPID-0 - The detector

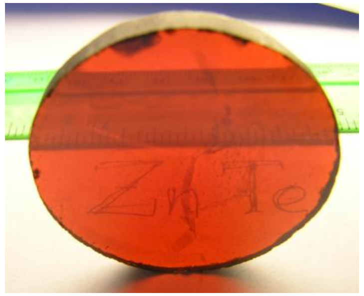


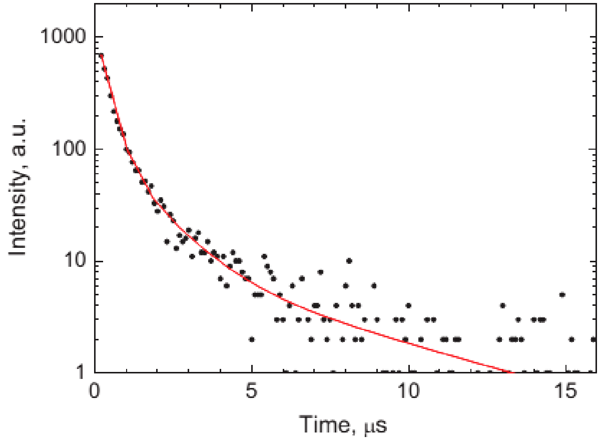
Previous NEMO limit $T_{1/2}^{0\nu}(^{82}Se) > 3.6 \cdot 10^{23}$ yr

ZnTe cryogenic scintillator

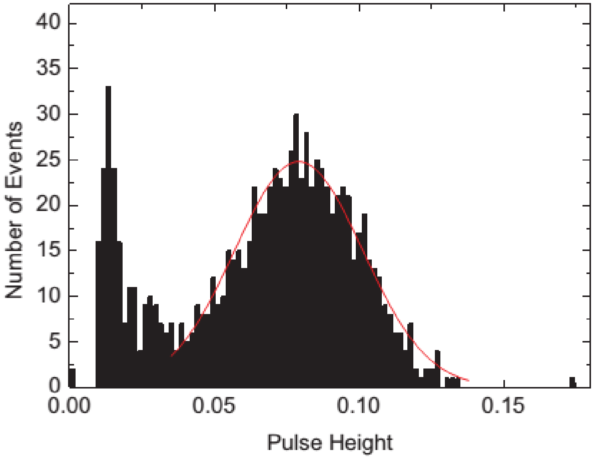


X-ray luminescence spectra of undoped ZnTe at T=120 K (1) 80 K (2), 40 K (3) and 10 K (4).

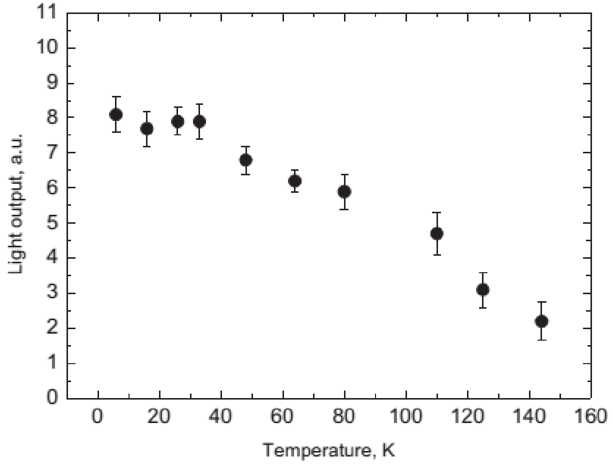




Scintillation decay curve of ZnTe (T=6 K). Excitation by α-particles from ^{241}Am (5.5 MeV). The best fit to using three exponentials with τ_1=0.3, τ_2=1.2 and τ_3=5.6 μs



Pulse height spectrum of scintillations excited by α-particles from ^{241}Am in ZnTe at T=6 K. The spectrum is fitted by Gaussian (red).



Temperature dependence of light output of ZnTe. Excitation by α-particles from ^{241}Am (5.5 MeV)

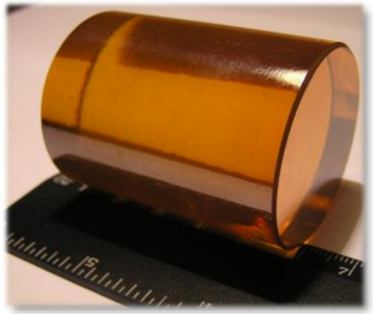
ISMA scintillation crystals to search for REP

- ZnSe 2β ^{64,70}Zn, ^{82}Se ; DM
- ZnTe 2β ^{64,70}Zn, ^{130}Te; DM
- $CdWO_4$ 2β ^{106,108,114,116}Cd, ^{180}W,^{186}W;DM
- $ZnWO_4$ 2β ^{64,70}Zn, ^{180}W,^{186}W; DM
- $^{ARCH}PbWO_4$ light guides, 2β ^{180}W,^{186}W; DM
- $^{ARCH}PbMoO_4$ 2β ^{100}Mo, DM
- $ZnMoO_4$ 2β ^{100}Mo, ^{64,70}Zn, DM
- Li_2MoO_4 2β ^{100}Mo, ^{64,70}Zn, DM
- $MgWO_4$ DM
- $ZnMgWO_4$ DM

$ZnWO_4$

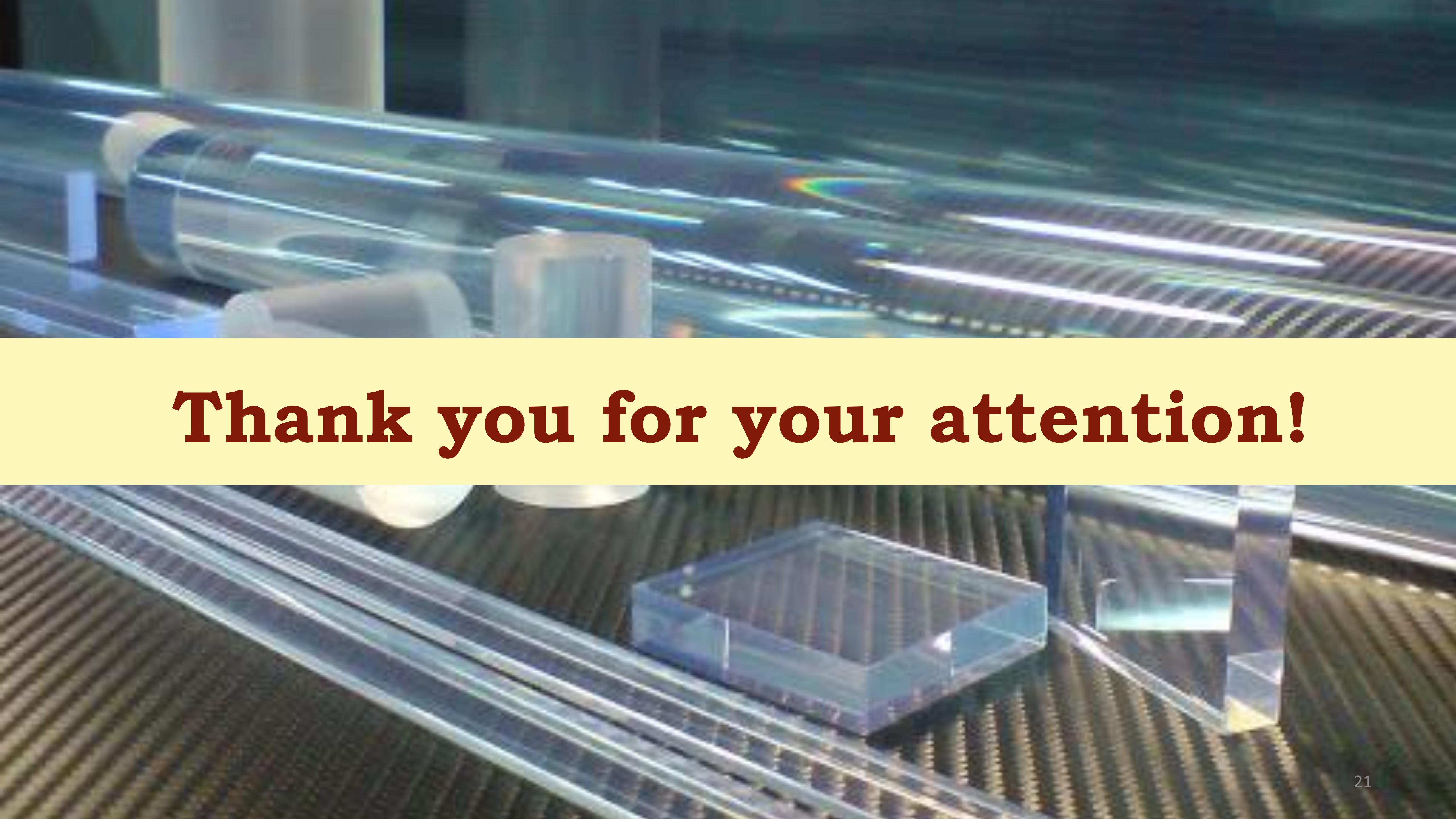
$PbWO_4$

$MgWO_4$



ZnSe

$ZnMoO_4$



Thank you for your attention!

$ZnWO_4$

monoclinic structure

ionic radius

Zn^{2+} - 0.074 nm

$MgWO_4$

monoclinic structure

ionic radius

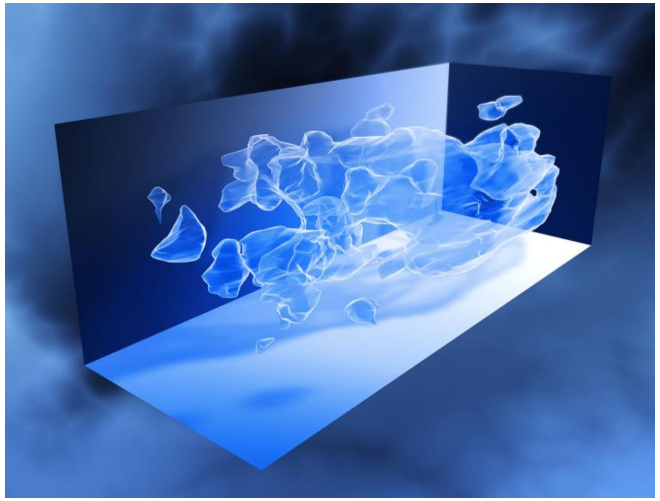
Mg^{2+} - 0.072 nm

continuous series of solid solutions of zinc and magnesium tungstate with different ratios of heavy zinc / light magnesium

Dark Matter

We cannot explain nature of 95% of *the Universe* mass

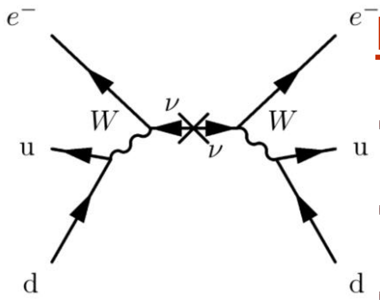
new kind of matter (new particles)



3D map of the large-scale distribution of dark matter, reconstructed from measurements of weak gravitational lensing with the Hubble Space Telescope.

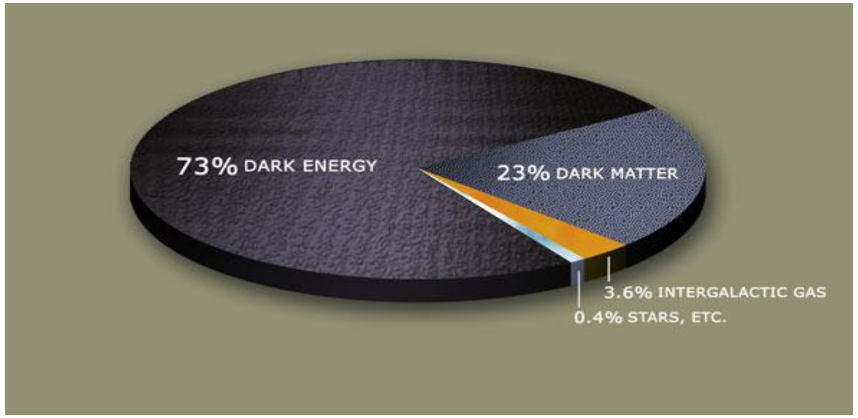
Popular candidates for the role of DM particles

- **Weakly Interacting Massive Particles (WIMP)**
- **Neutrino**



Double beta decay

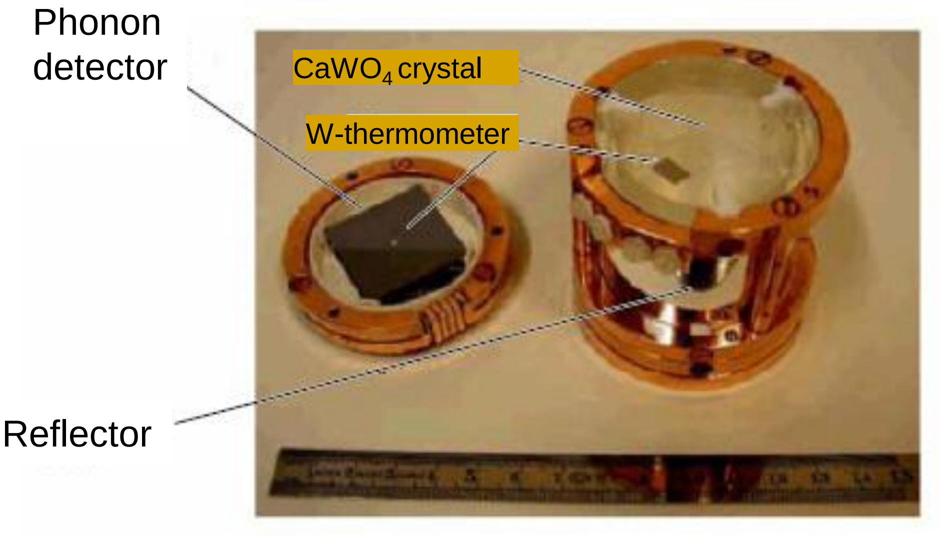
- **Nature of neutrino**
- **Neutrino mass**
- **Lepton number conservation**

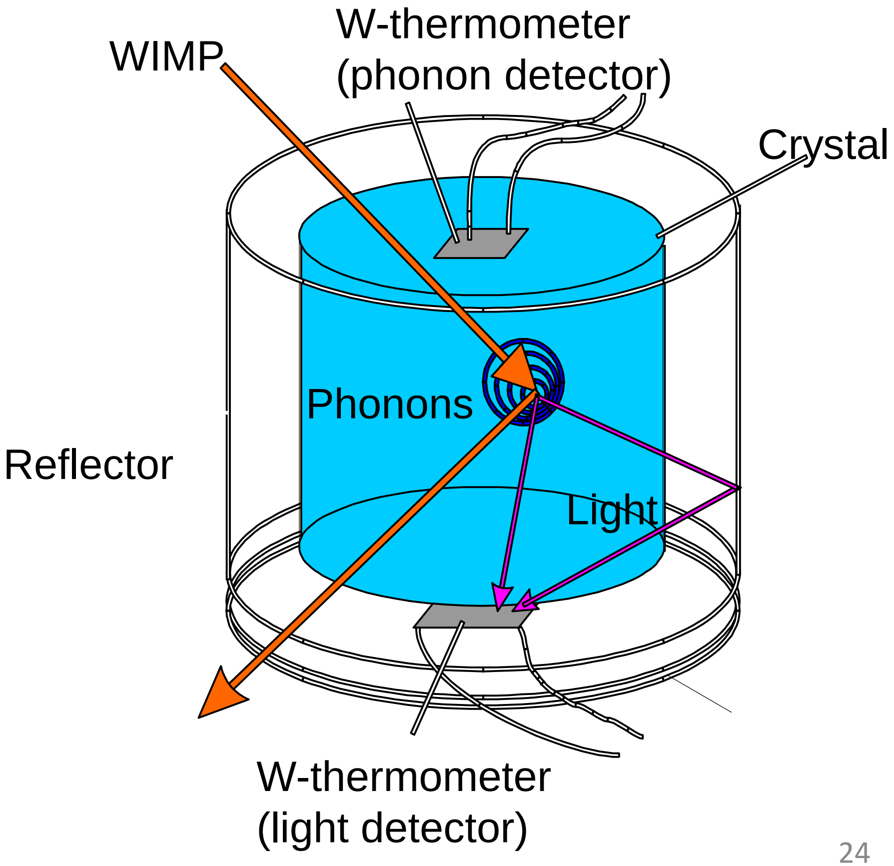


Combined cryogenic detectors

- The energy threshold of the recoil energy of -10 keV.
- High energy resolution, comparable with germanium detectors.
- Effective separation of events from the γ-rays (β–, α–particles) and the recoil nucleus.

CRESST-II detector





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