$ZnMoO_4$ purification and crystallization in Novosibirsk

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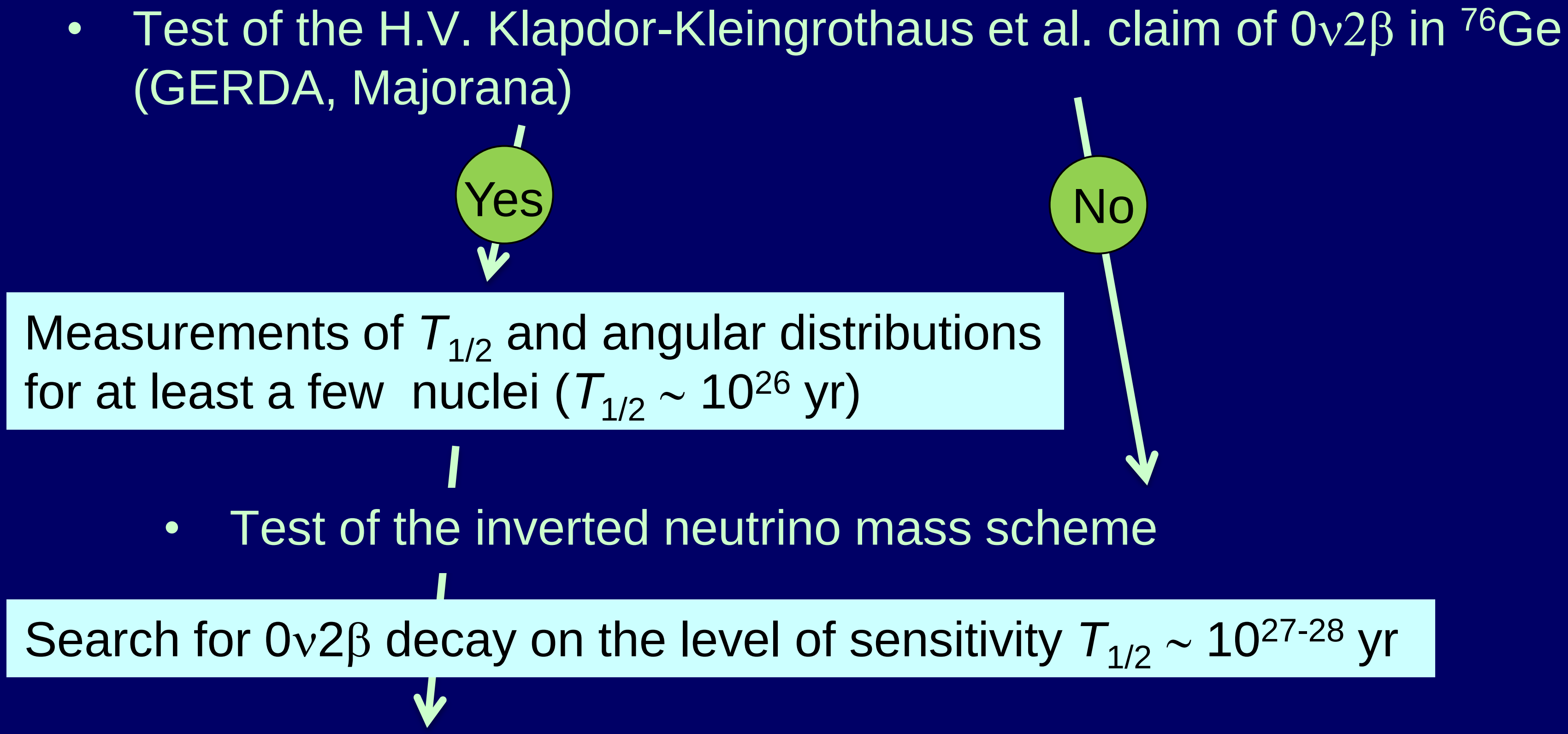
Prospects for 2β experiments



- Test of the H.V. Klapdor-Kleingrothaus et al. claim of 0ν2β in ^{76}Ge (GERDA, Majorana)

Measurements of $T_{1/2}$ and angular distributions for at least a few nuclei ($T_{1/2} \sim 10^{26}$ yr)

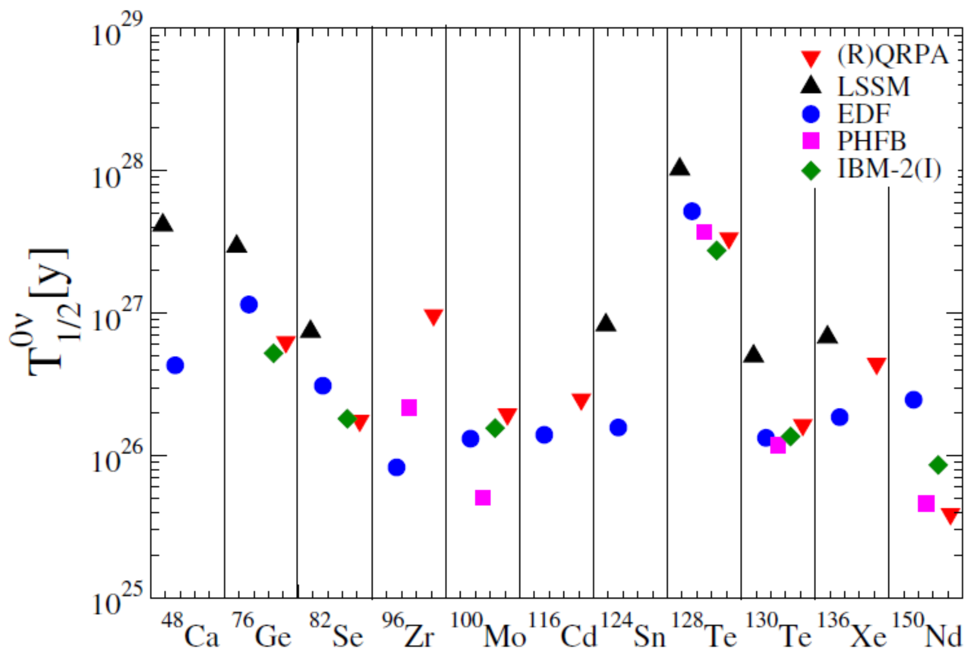
- Test of the inverted neutrino mass scheme

Search for 0ν2β decay on the level of sensitivity $T_{1/2} \sim 10^{27\text{-}28}$ yr

- Investigation of 2ε, $\varepsilon\beta^+$ and $2\beta^+$ processes (a possibility to distinguish between the ν mass and the right handed current mechanisms)
- Precise measurement of 2ν channels

Test of the neutrino mass hierarchy

Theoretical calculations of $T_{1/2}$ for $\langle m_\nu \rangle = 0.05$ eV [1]



To cover the inverted hierarchy region, one needs a sensitivity:
$\langle m_\nu \rangle \sim 0.02$ eV $\rightarrow$ $T_{1/2} \sim 10^{27} - 10^{28}$ yr

[1] J.D.Vergados, H.Ejiri, F.Simkovic, Rep. Prog. Phys. 75 (2012) 106301

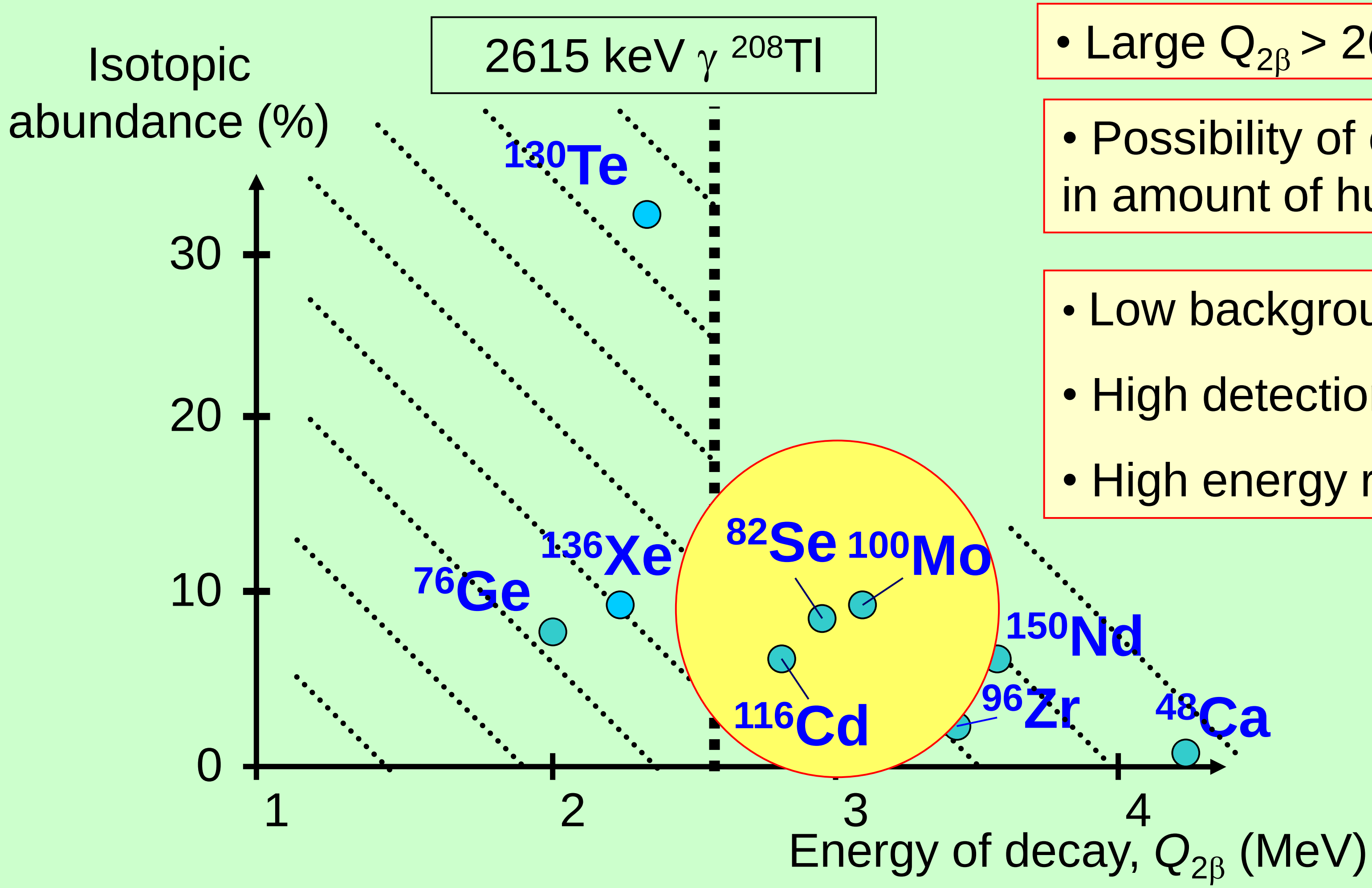
What does it mean $T_{1/2} \sim 10^{27} - 10^{28}$ yr ?

Nucleus	$T_{1/2}$ to reach $\langle m_\nu \rangle = 0.02$ eV [1]	Detector	Number of 2β nuclei in 1 ton detector	Number of decays over 5 yr
^{48}Ca	$(3 - 28) \times 10^{27}$ yr	^{48}CaF$_2$ (20%)	1.4×10^{27}	0.2 – 1.9
^{76}Ge	$(3 - 17) \times 10^{27}$ yr	HP ^{76}Ge	7.9×10^{27}	1.6 – 9
^{82}Se	$(1 - 4) \times 10^{27}$ yr	Zn^{82}Se	4.1×10^{27}	3 – 13
^{100}Mo	$(0.3 - 1.5) \times 10^{27}$ yr	Zn^{100}MoO$_4$	2.6×10^{27}	6 – 30
		^{40}Ca^{100}MoO$_4$	3.0×10^{27}	4 – 34
^{116}Cd	$(0.8 - 1.3) \times 10^{27}$ yr	^{116}CdWO$_4$	1.7×10^{27}	4 – 7
^{130}Te	$(0.7 - 3) \times 10^{27}$ yr	^{130}TeO$_2$	3.8×10^{27}	4 – 18
^{136}Xe	$(1 - 4) \times 10^{27}$ yr	^{136}Xe	4.4×10^{27}	4 – 14

[1] Table 3 in J.D.Vergados, H.Ejiri, F.Simkovic, Rep. Prog. Phys. 75 (2012) 106301

The most “promising” 2β nuclei

from the point of view of experiment



- Large $Q_{2\beta} > 2615$ keV

- Possibility of enrichment in amount of hundreds kg

- Low background
- High detection efficiency
- High energy resolution *)

***) Pure energy resolution is still acceptable if one give a *limit* on $0\nu2\beta$ decay, while it is not a case if one claim *detection* of the process**

LUMINEU in a nutshell

Luminescent **U**nderground **M**olybdenum **I**nvestigation for **NEU**trino mass and nature

Expérience souterraine avec détecteurs luminescents de molybdate de zinc pour l'étude de la masse et la nature des neutrinos

① Set the bases for a **next-generation neutrinoless double-beta** decay experiment

- $ZnMoO_4$ crystal production
- Temperature sensor production and optimization
- Light detector development
- Pilot experiment with enriched material

But also:

② Possible implications on **the direct detection of dark matter** (especially for low-mass WIMPs)

Funded by **ANR** in France (Agence National de la Recherche)
Collaboration : **CNRS-Orsay, CEA-Saclay, IAS-Orsay, ICMCB Bordeaux, INR Kiev, NIIC Novosibirsk ; Heidelberg University** is joining. In total, ~ 40 participants.
Start: October 1st, 2012 – duration: 4 years

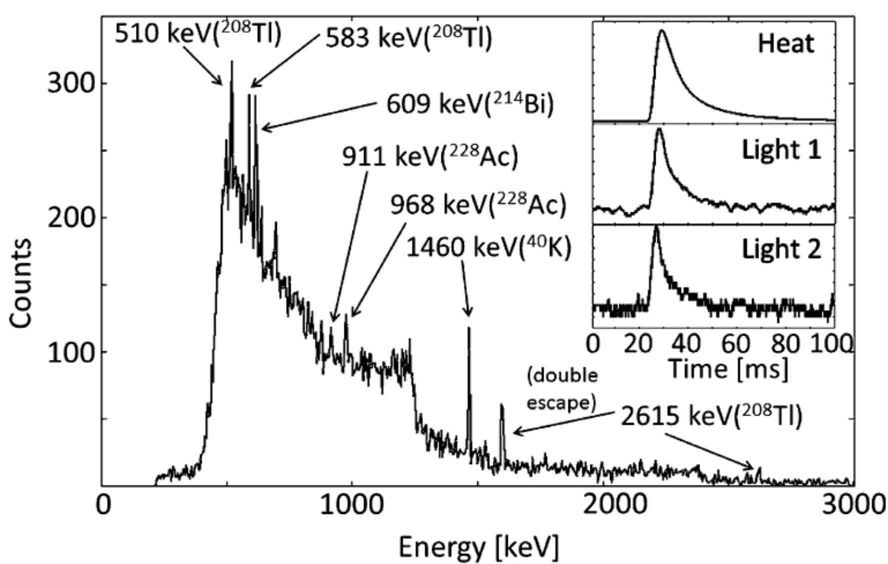
Courtesy Andrea Giuliani

Properties of ZnMoO$_4$ crystals

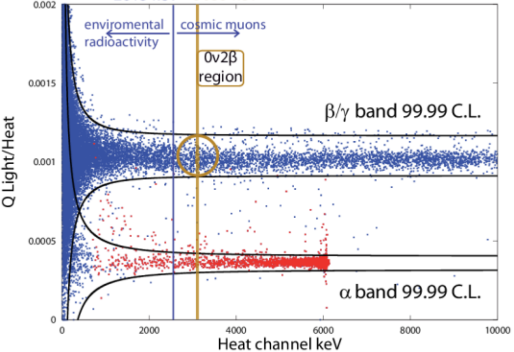
Property	**Value**	**Measurements**	**Reference**
Density (g/cm^3)	4.3		[1]
Melting point (° C)	1003 ± 5		[1]
Structural type	Triclinic, *P*1		[1, 2]
Cleavage plane	Weak (001)		[1]
Hardness Mohs scale	3.5		[3]
Index of refraction	1.89 – 1.96		[3]
Wavelength of emission maximum (nm)	605 585 625	SR 6.5 eV, 10 K X ray excitation, 8 K X ray excitation, 8 K	[1] [4] [3]
Scintillation decay time (μs)	1.3, 16, 150 3.9	SR 6.5 eV, 80 K SR 5.5 eV, 300 K	[5] [6]

[1] L.I.Ivleva *et al*., Crystallog. Rep. 53 (2008) 1087
[2] W.Reichelt *et al*., Z. Anorg. Allg. Chem. 626 (2000) 2020
[3] D.M.Chernyak et al., in review in NIMA
[4] L.L.Nagornaya *et al*., IEEE Trans. Nucl. Sci. 56 (2009) 2513
[5] V.B. Mikhailik et al., Nucl. Instr. Meth. A 562 (2006) 513
[6] D. Spassky et al., Phys. Status Solidi A 206 (2009) 1579

$ZnMoO_4$ scintillating bolometers



Chain	Activity (mBq/kg)	
	[1]	[2]
^{226}Ra	< 0.8	= 0.027(6)
^{228}Th	< 0.8	< 0.006



- **High energy resolution 3.8 keV at 2615 keV (0.15%)**
- **Estimated background is a few counts / yr at $Q_{2\beta}$ in 1 ton detector (the main background is expected to be from random coincidence of 2ν2β events [3])**

Sensitivity for 5 yr 800 kg $Zn^{100}MoO_4$: $T_{1/2} \approx 10^{27}$ yr → $\langle m_\nu \rangle \sim 0.013 - 0.05$ eV [4]

[1] D.M. Chernyak *et al.*, submitted to NIMA; [2] J.W.Beeman *et al.*, Eur. Phys. J. C 72 (2012) 2142
[3] D.M. Chernyak *et al.*, Eur. Phys. J. C 72 (2012) 1989; [4] J.W. Beeman *et al.*, PLB 710 (2012) 318

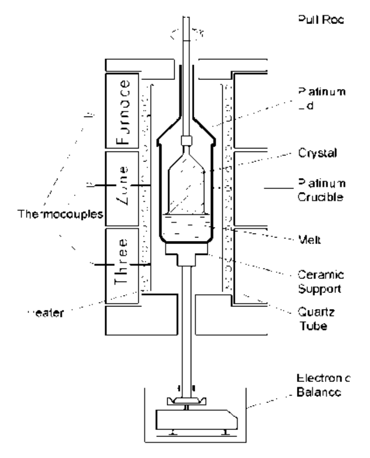
R&D of $Zn^{100}MoO_4$ crystal scintillators

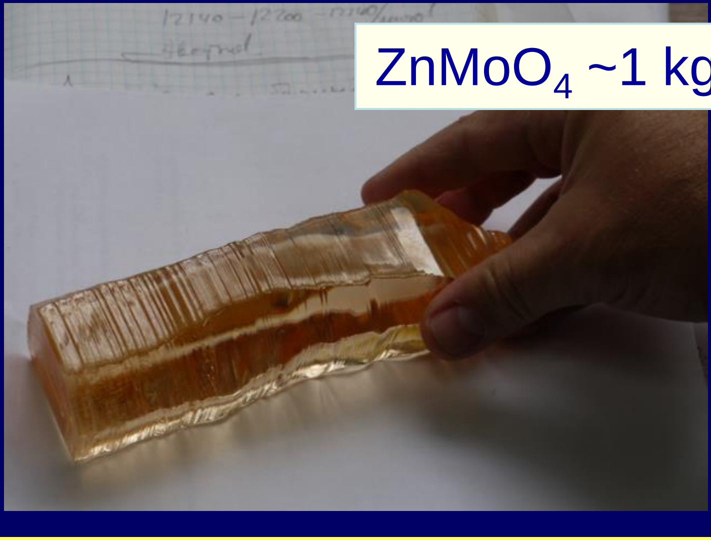
- Deep purification of Mo
- Recovery of Mo from $ZnMoO_4$
- Crystal growth
- Scintillation elements production
- Tests of produced crystal samples

Low-Thermal-Gradient Czochralski technique

Nikolaev Institute of Inorganic Chemistry, Novosibirsk, Russia

Low-Thermal-Gradient Czochralski (LTG-C) [1]





$ZnMoO_4$ ~1 kg

	standard	LTG-C
Output	25-30%	up to 90% *)
Quality		typically higher *)
Radiopurity		expected better
Loses of powder	2-3%	<0.3% *)

***) Achieved for $^{106}CdWO_4$ and $^{116}CdWO_4$ [2, 3]**

[1] A.A. Pavlyuk *et al.*, Proc. *APSAM*-92, April 26–29, Shanghai, China (1992)
[2] P. Belli *et al.*, NIMA 615 (2010) 301; [3] A. Barabash *et al.*, JINST 6 (2011) P08011

Status of $Zn^{100}MoO_4$ R&D

First LUMINEU $ZnMoO_4$ samples were delivered in June 2013 (in the frame of the 1st contract):

- 2 samples ∅ 20 × 40 mm
- 2 samples ∅ 35 × 40 mm

Tests:

- Bolometric properties
- Radioactive contamination
- Optical and luminescence properties
- Diamagnetic properties
- Debye temperature
- Segregation of impurities
- Screening of radioactive contamination of polishing materials

next steps

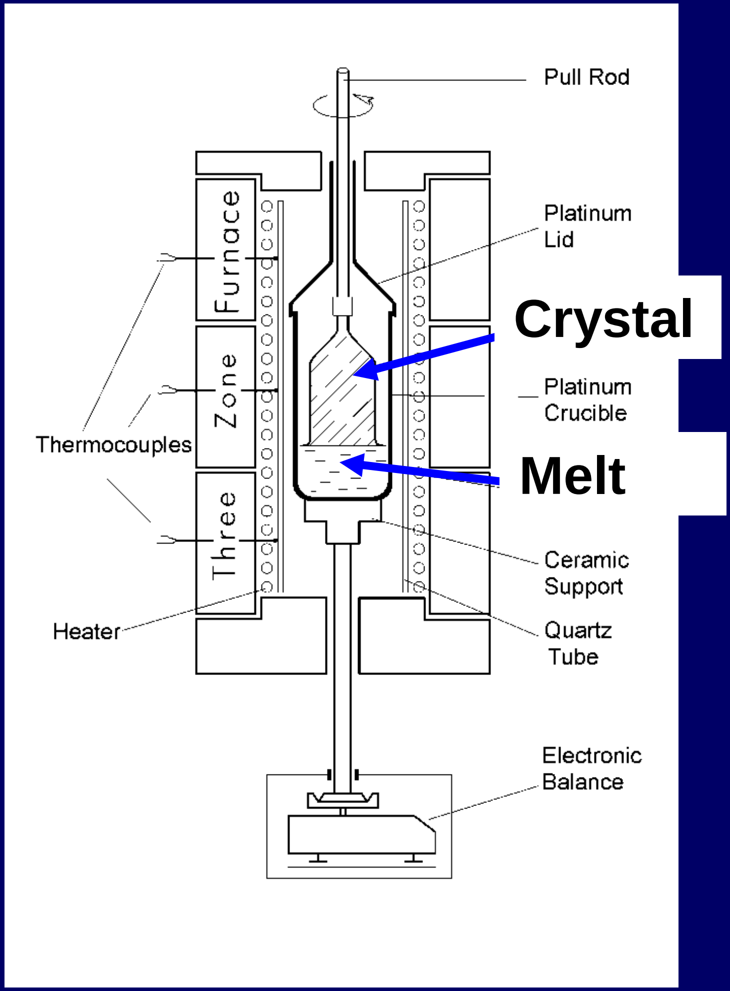
(to be realized in the frame of the next 3 contracts)

- Production of "small" (≈ ∅ 20 mm) enriched $Zn^{100}MoO_4$ crystal(s) from ≈180 g of contaminated $^{100}MoO_3$ *)
- Growth of $ZnMoO_4$ from deeply purified Mo
- Recrystallization
- Increase diameter (goal ~ 6 cm)
- Growth of "large" (≈ ∅ 40 - 50 mm) enriched $Zn^{100}MoO_4$ crystal(s) from ≈ 1 kg of ^{100}Mo

*) rest after wet chemistry purification of $^{100}MoO_3$ for the ARMONIA experiment [1]:
a few mBq/kg of ^{228}Th and ^{226}Ra, 0.3 Bq/kg ^{40}K, 20 mBq/kg of ^{137}Cs)

[1] P.Belli *et al.*, Nucl. Phys. A 846(2010)143

Segregation of radioactive elements in crystals



Segregation of impurities

$$K = C_S / C_L,$$

where K is segreagation coefficient, C_S is concentration of impurity in solid phase (crystal), C_L is concentration of impurity in liquid phase (melt),

If $K < 1$, recrystallization could improve radiopurity of the crystal

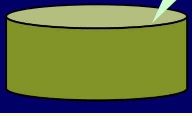
Possibility to improve radiopurity by recrystallization (on example of ^{116}CdWO$_4$)

Activity of ^{228}Th (mBq/kg)

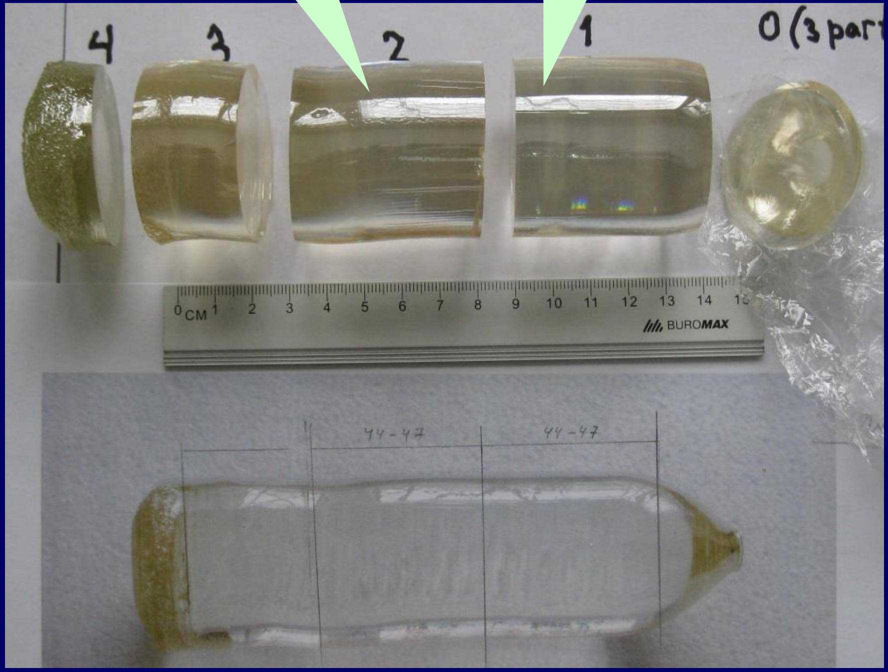
10(2)

0.07(1)

0.04(1)



rest of the melt



Nuclide	Activity (mBq/kg)	
	Crystal	Rest of melt
^{40}K	<1	27(11)
^{226}Ra	<0.005	64(4)
^{228}Th	0.04 – 0.07	10(2)

We expect to reduce K, Th, U and Ra contamination by recrystallization

D.V. Poda *et al.*, Radiat. Meas., DOI 10.1016/j.radmeas.2013.02.017
F.A. Danevich *et al.*, to be published in LRT 2013 proceedings

conclusions

- The next generation 2β experiments call for large mass detectors (~100 - 1000 kg) with challenging characteristics: containing certain elements → isotopically enriched, very low radioactive background, high energy resolution (< 1%), long time operation (~ 10 yr)
- Crystal scintillators, in particular $ZnMoO_4$, can meet these requirements
- Production of high quality low radioactive crystal scintillators from enriched isotopes requires a special extended R&D
- Purification of raw materials is an important issue to obtain high quality radio-pure $ZnMoO_4$ and $Zn^{100}MoO_4$ crystals
- Recrystallization could be a way to improve further the $ZnMoO_4$ radiopurity
- R&D of methods to recover Mo from $ZnMoO_4$ scraps is in progress

RPSCINT 2013

International Workshop on Radiopure Scintillators

September 17-20, 2013

National Academy of Sciences of Ukraine
Institute for Nuclear Research, Kyiv, Ukraine

The workshop is dedicated to the 70-th anniversary of Yuri Zdesenko

TOPICS

- radiopure scintillators in nuclear and astroparticle physics
- requirements of low-count rate experiments regarding radiopurity and scintillation properties
- radioactive contamination of scintillation materials
- selection and screening of input materials
- instruments and methods to test radioactive contamination of materials and scintillators
- purification of materials and preparation of raw compounds
- crystal growing, annealing and handling
- test of scintillators including scintillation, optical, luminescence low-background and low-temperature measurements
- search for and development of new scintillating materials

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