

Examination of natural and enriched materials for ZnSe crystal production

F. Orio – INFN sez. Roma I
on behalf of LUCIFER collaboration

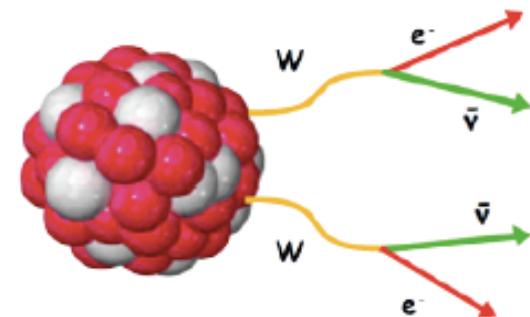
4th ISOTTA general meeting – CSNSM ORSAY – December 1st, 2014

Double-Beta Decay

It is a very rare nuclear decay:
 $(A, Z) \rightarrow (A, Z+2) + 2e^- + (2\bar{\nu}_e)$

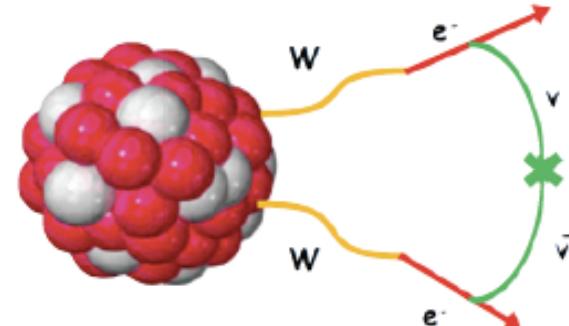
$2\nu\beta\beta$

- Second order SM weak process
- Rarest decay ever observed: $T_{1/2} \sim 10^{19} - 10^{21}$ y

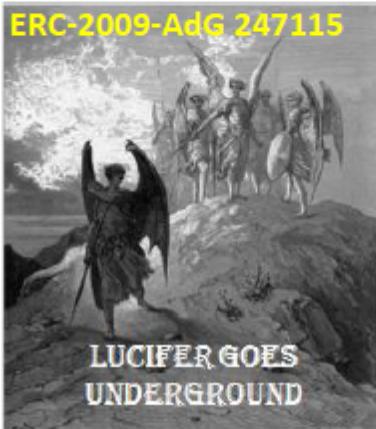


$0\nu\beta\beta$

- Not allowed by the Standard Model ($\Delta L=2$)
- Decay never observed: $T_{1/2} > 10^{22} - 10^{25}$ y
- Possible only if neutrinos are Majorana particles



LUCIFER Low-background Underground Cryogenics Installation For Elusive Rates



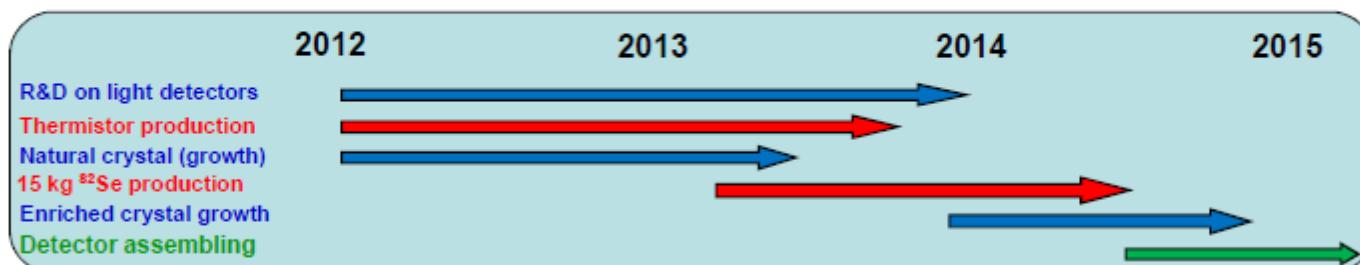
Principal Investigator: Prof. Fernando Ferroni
Co-Investigator : Prof. Andrea Giuliani

isotope: ^{82}Se , ^{100}Mo , ^{116}Cd
material: ZnSe , ZnMoO_4 , CdWO_4
technique: scintillating bolometer

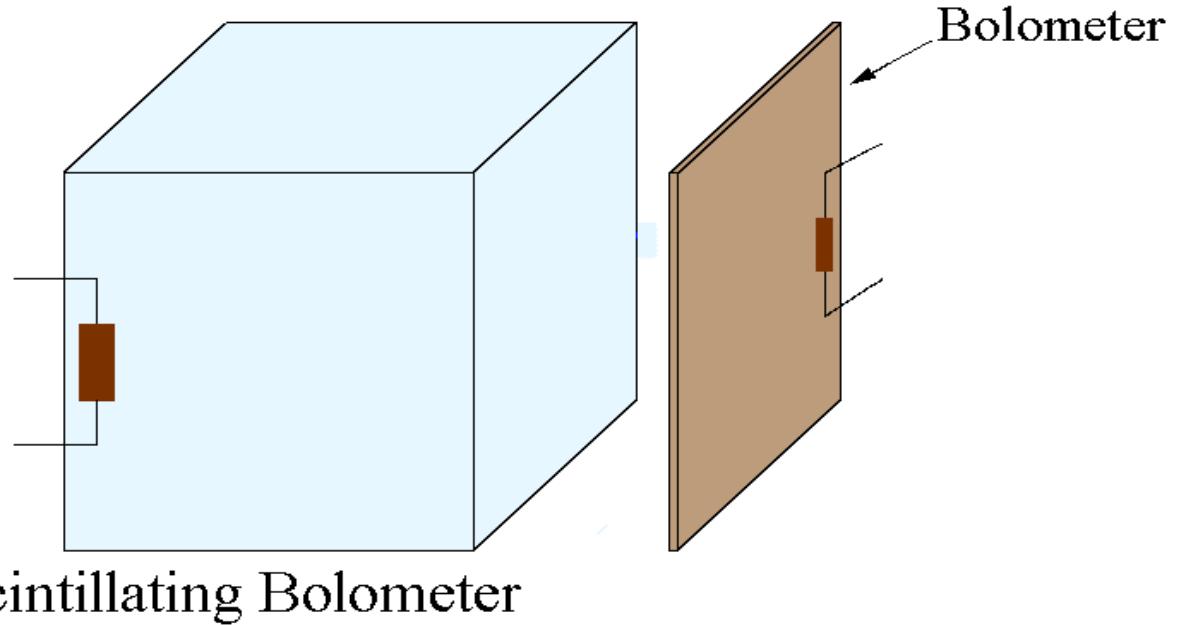
array of 36÷44 enriched (95%) Zn^{82}Se crystals
total ^{82}Se nuclei: $(7.1 \div 8.9) 10^{25}$
expected background in the ROI (2995 keV): $(3 \div 6) 10^{-3} \text{ c/keV/kg/y}$

demonstrator

LUCIFER timeline



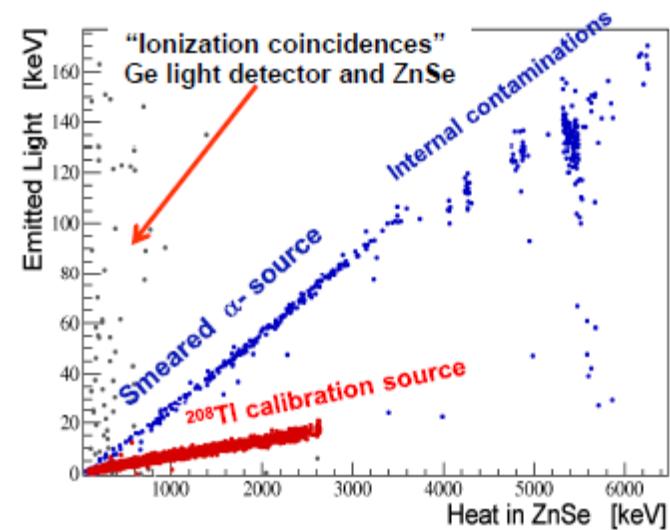
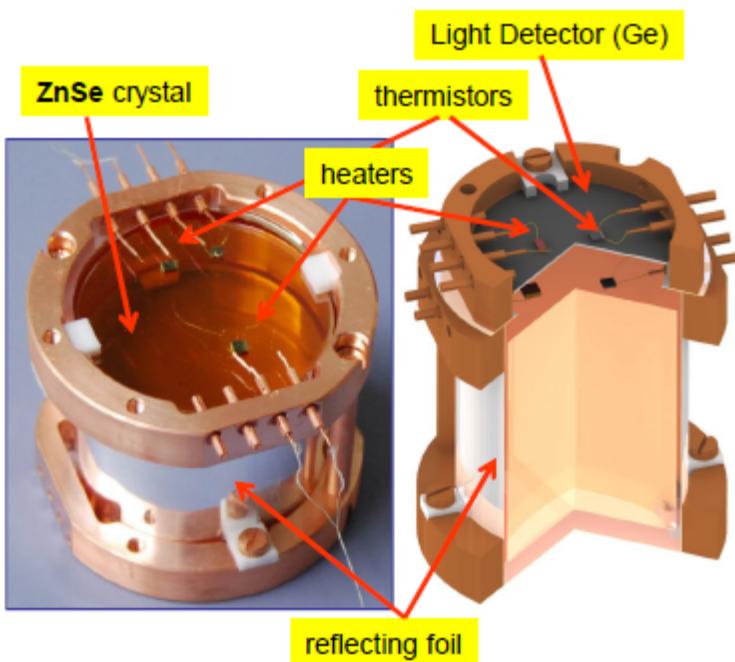
Working principle of scintillating bolometers



First measurement of light and heat in a
bolometer made in 1992 at INFN Milano

A. Alessandrello et al., "Development of a
thermal scintillating detector for double
beta decay of ^{48}Ca " Nucl. Phys. B -
Proceedings Supplements 1992 Vol: 28.

Typical mounting & scatter plot for ZnSe crystal



$$QF_{\alpha} > 1$$

Requirements to raw material for ZnSe crystal production

High performance of ZnSe crystals as scintillating bolometer strongly depends on:

- **High perfection of crystalline structure** (*FWHM & Signal amplitude in Heat channel*)
- **High Light Yield** (*FWHM & Signal amplitude in Light channel, particle discrimination*)

Chemical purity

K, Ti, V, Cr, Mn, **Fe**, Co, Ni $< 1 \times 10^{-6}$ g/g

Mg, Bi, Pb, Sn, Al, Cu, Cd $< 5 \times 10^{-6}$ g/g

U, Th $<< 1 \times 10^{-9}$ g/g

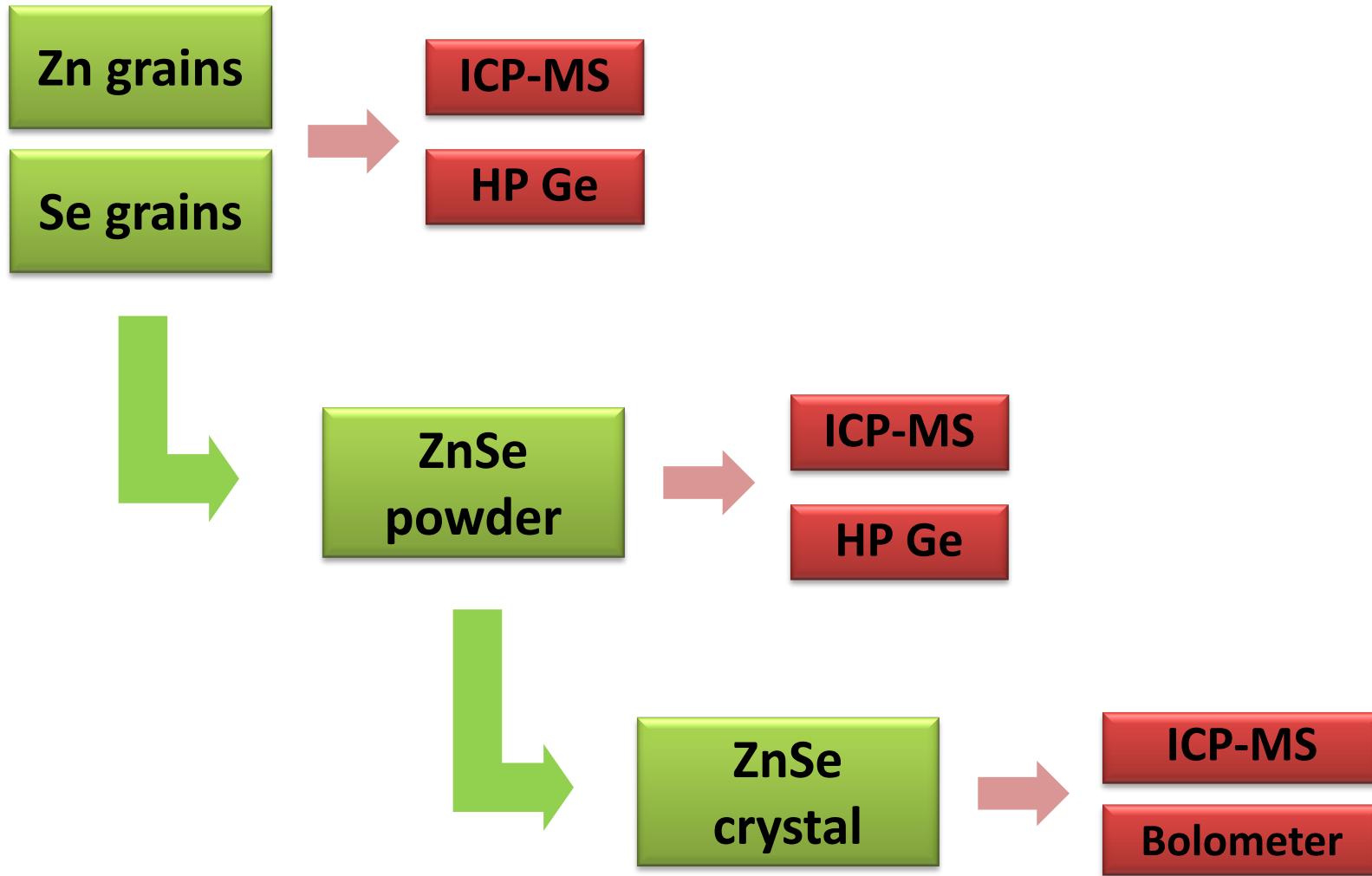
Radiopurity

A(⁴⁰K, U/Th& daughters) $<$ mBq/kg

no ¹³⁷Cs, ⁹⁰Sr, ...

minimization of cosmogenic nuclides activity

Examination scheme of samples for ZnSe crystal production



Chemical purity of Zn, Se & ZnSe powder samples

(concentration present in unit of 10^{-9} g/g)

| Element | Se grains | Zn grains | ZnSe powder |
|---------|-----------|-----------|-------------|
| Na | < 102 | < 46 | < 42 |
| Mg | < 3380 | < 1530 | < 1390 |
| Si | < 17000 | < 15000 | 4200 |
| S | < 34000 | < 15000 | < 14000 |
| V | < 7 | < 3 | < 3 |
| Cr | < 33.8 | 4.6 | 1.1 |
| Mn | < 68 | < 31 | < 28 |
| Fe | < 337.9 | < 152.9 | 139 |
| Co | < 6.8 | < 3 | < 2.8 |
| Ni | < 34 | < 15 | < 7 |
| Cu | < 27 | 153 | 63 |
| As | < 3400 | < 31 | < 1400 |
| Y | < 3.4 | < 1.5 | < 1.4 |
| Mo | < 1351 | 3 | < 883 |
| Cd | 500 | 500 | 700 |
| Sb | 6.8 | < 1.5 | 2.8 |
| Te | 743 | 382 | 695 |
| W | 10 | < 1.5 | 7 |
| Hg | 574 | 20 | 14 |
| Tl | < 338 | 15 | 14 |
| Pb | 17 | 184 | 28 |
| Bi | 3.4 | 1.5 | 0.3 |
| Th | < 0.14 | < 0.015 | < 0.014 |
| U | < 0.14 | < 0.06 | < 0.06 |

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(concentration present in unit of 10^{-9} g/g)

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Quartz reactor
for ZnSe synthesis

Reduction during
ZnSe synthesis

Traces from
CdTe, CdZnTe,
CdHgTe production

Radiopurity of natural Zn, Se & ZnSe powder samples

(activity present in unit of mBq/kg)

| Chain | Nuclide | Se grains | Zn grains | ZnSe powder |
|-------|--------------------|----------------|------------------|-------------|
| 232Th | ²²⁸ Ra | < 0.67 | < 0.65 | |
| | ²²⁸ Th | < 0.56 | < 0.81 | |
| | | | | |
| 238U | ²²⁶ Ra | < 0.51 | < 0.42 | |
| | ²³⁴ Th | < 27 | < 36 | |
| | ^{234m} Pa | < 14 | < 19 | |
| 235U | ²³⁵ U | < 0.99 | < 0.48 | |
| | | | | |
| | ⁴⁰ K | < 14 | < 10 | |
| | | | | |
| | ⁶⁰ Co | < 0.2 | < 0.082 | |
| | | | | |
| | ¹³⁷ Cs | 0.5±0.2 | < 0.50 | |
| | | | | |
| | ⁷⁵ Se | 8.2±0.7 | - | |
| | | | | |
| | ⁵⁴ Mn | - | < 0.28 | |
| | | | | |
| | ⁵⁶ Co | - | 0.33±0.14 | |
| | | | | |
| | ⁵⁸ Co | - | 0.43±0.16 | |
| | ⁶⁵ Zn | - | 7.6±1.0 | |
| | | | | |

Radiopurity of natural Zn, Se & ZnSe powder samples

(activity present in unit of mBq/kg)

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|-------------------|--------------------|------------------|----------------|-------------|
| 232Th | ²²⁸ Ra | < 0.67 | < 0.65 | < 2.3 |
| | ²²⁸ Th | < 0.56 | < 0.81 | < 2.1 |
| 238U | ²²⁶ Ra | < 0.51 | < 0.42 | < 3.8 |
| | ²³⁴ Th | < 27 | < 36 | < 120 |
| | ^{234m} Pa | < 14 | < 19 | < 140 |
| 235U | ²³⁵ U | < 0.99 | < 0.48 | < 2.0 |
| | ⁴⁰ K | < 14 | < 10 | < 22 |
| | ⁶⁰ Co | < 0.2 | < 0.082 | < 0.55 |
| ¹³⁷ Cs | 0.5±0.2 | < 0.50 | 3.4±1.0 | |
| | 8.2±0.7 | - | 4.5±1.0 | |
| ⁵⁴ Mn | - | < 0.28 | - | |
| | 0.33±0.14 | | - | |
| ⁵⁶ Co | - | 0.43±0.16 | - | |
| | 7.6±1.0 | | - | |
| ⁶⁵ Zn | - | 7.6±1.0 | 2.0±1.0 | |
| | | | | |

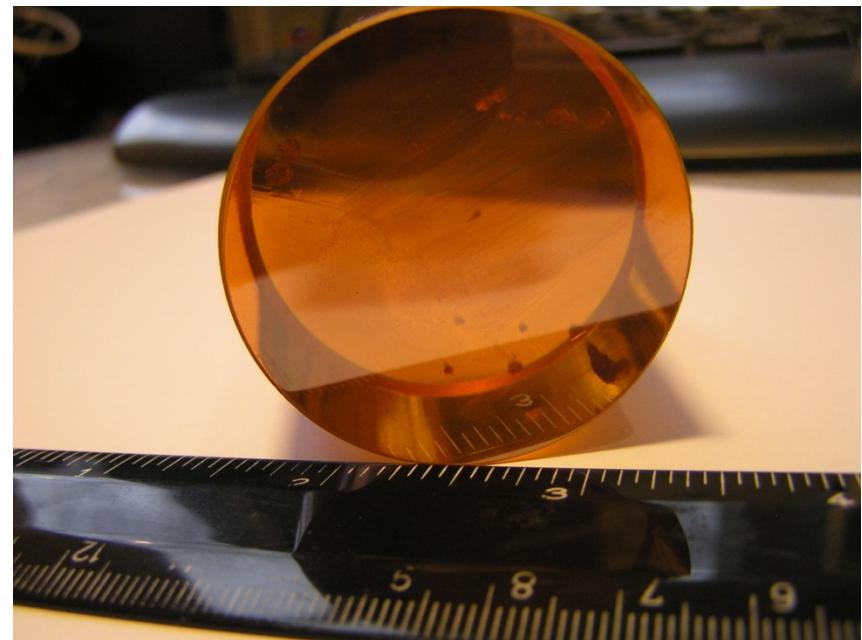
Contamination during
ZnSe synthesis

Reduction in
accordance with Se
mass fraction (54.7%)

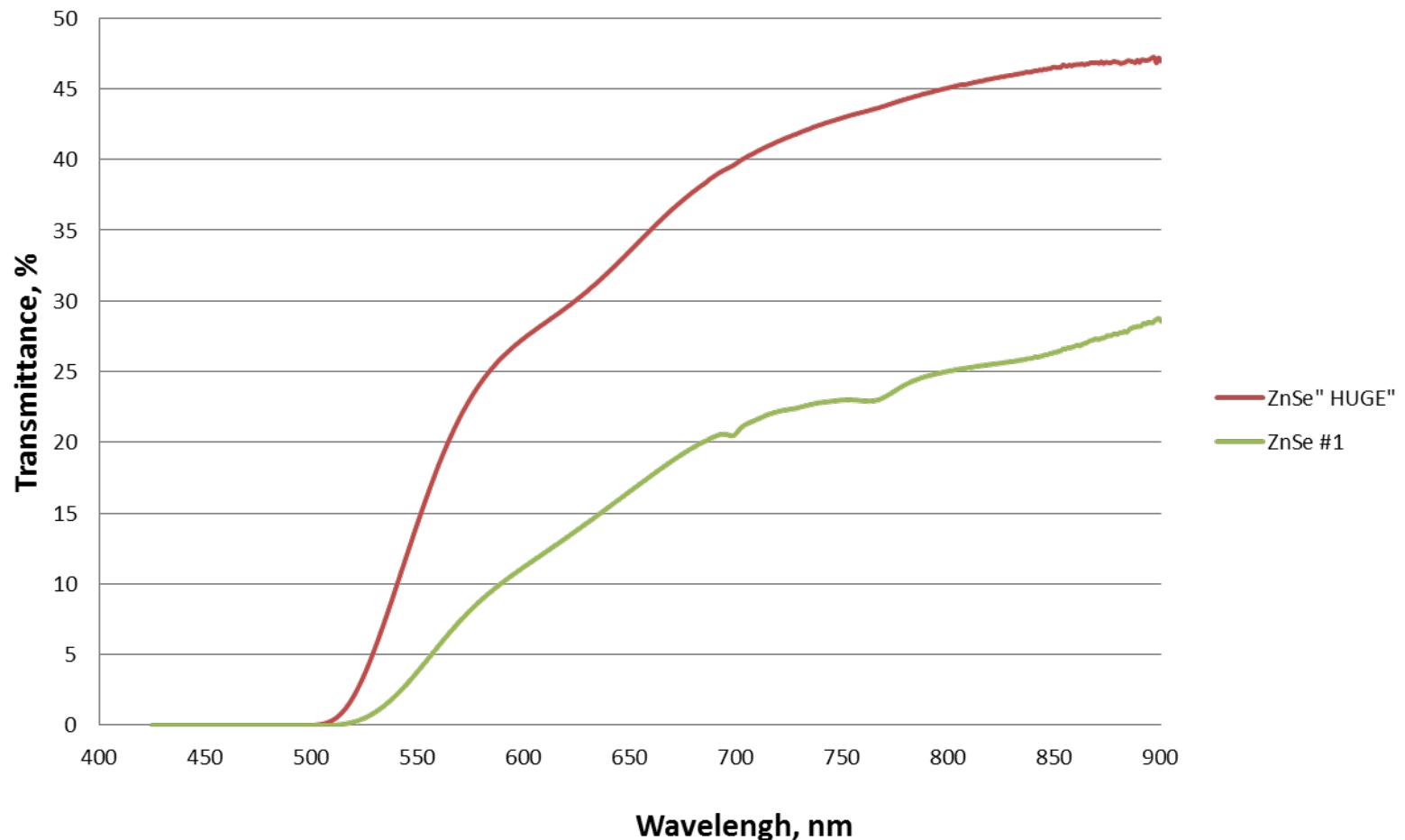
Reduction/not
enough statistics

More reduction than
expected based on
Zn mass fraction
(45.3% - 3.5 mBq/kg)

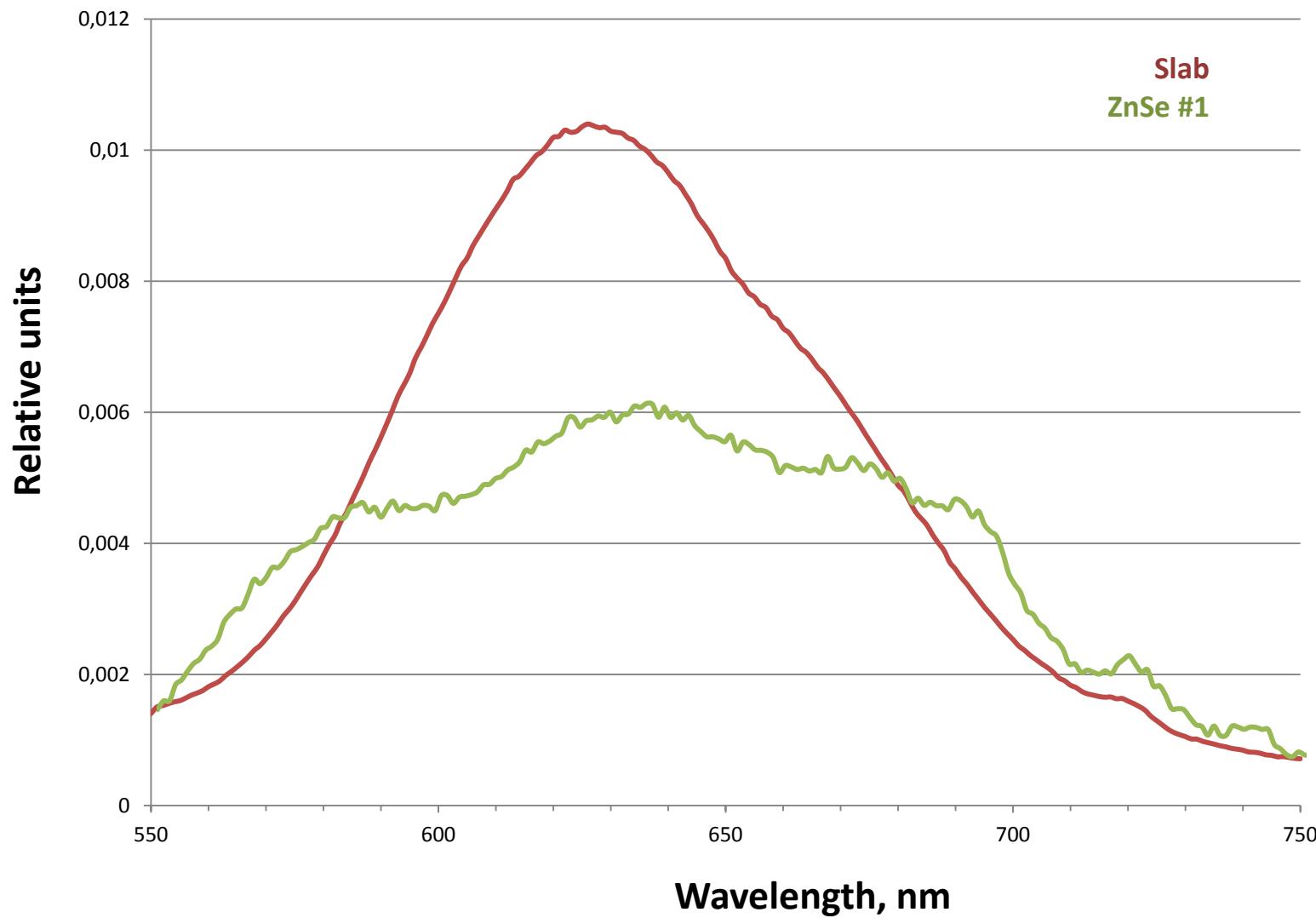
ZnSe #1 crystal, which was produced from analyzed ZnSe powder



Transmittance of ZnSe #1 crystal and ZnSe «Huge» (the best crystal) in comparison



Emission spectra @ excitation 480 nm



Chemical purity of HP Zn & ^{82}Se metal samples

(concentration present in unit of 10^{-9} g/g)

| Element | ^{82}Se grains (conv. 38) | HP Zn grains |
|---------|---------------------------------------|--------------|
| Na | < 1860 | < 850 |
| Mg | < 186 | < 38 |
| Si | < 9090 | < 8000 |
| S | 163600 | < 9000 |
| V | < 4 | < 9 |
| Cr | 18 | < 90 |
| Mn | < 37 | < 90 |
| Fe | < 91 | < 180 |
| Co | < 2 | < 10 |
| Ni | < 91 | < 90 |
| Cu | < 93 | 110 |
| As | < 91 | < 10 |
| Y | < 3 | < 1.5 |
| Mo | 9 | 4 |
| Cd | 9 | 4300 |
| Sb | < 4 | < 4 |
| Te | 840 | < 90 |
| W | 21 | < 10 |
| Hg | < 4 | < 2 |
| Tl | < 40 | 180 |
| Pb | < 9 | 290 |
| Bi | < 1 | < 2 |
| Th | < 0.9 | < 0.2 |
| U | < 0.9 | < 0.2 |

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| Cr | 18 | < 90 |
| Mn | < 37 | < 90 |
| Fe | < 91 | < 180 |
| Co | < 2 | < 10 |
| Ni | < 91 | < 90 |
| Cu | < 93 | 110 |
| As | < 91 | < 10 |
| Y | < 3 | < 1.5 |
| Mo | 9 | 4 |
| Cd | 9 | 4300 |
| Sb | < 4 | < 4 |
| Te | 840 | < 90 |
| W | 21 | < 10 |
| Hg | < 4 | < 2 |
| Tl | < 40 | 180 |
| Pb | < 9 | 290 |
| Bi | < 1 | < 2 |
| Th | < 0.9 | < 0.2 |
| U | < 0.9 | < 0.2 |

Contamination during ^{82}Se conversion from $^{82}\text{SeF}_6$ gas
!!! Further purification by annealing in H_2 flow before crystal growth

Residue in Zn due to similar chemical properties
 $A(^{113}\text{Cd}) = 8 \mu\text{Bq/kg}$

Residue in Se due to similar chemical properties

Residue due to instrumentation memory effect

Radiopurity of HP Zn & ^{82}Se metal samples

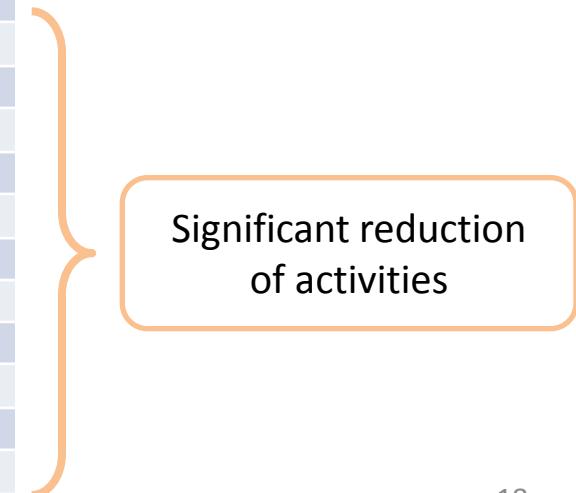
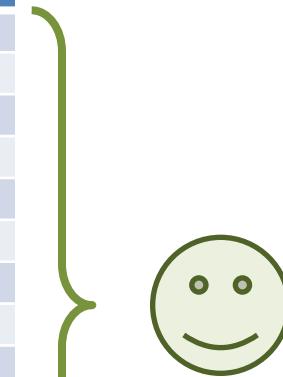
(activity present in unit of mBq/kg)

| Chain | Nuclide | ^{82}Se grains | HP Zn grains |
|-------------------|---------------------------|-----------------------------------|-----------------------------------|
| ^{232}Th | ^{228}Ra | < 0.082 | < 0.095 |
| | ^{228}Th | < 0.27 | < 0.036 |
| | | | |
| ^{238}U | ^{226}Ra | < 0.3 | < 0.066 |
| | ^{234}Th | < 6.9 | < 6.2 |
| | $^{234\text{m}}\text{Pa}$ | < 5.3 | < 4.7 |
| ^{235}U | ^{235}U | < 0.17 | < 0.091 |
| | | | |
| | ^{40}K | < 1.1 | < 0.38 |
| | | | |
| | ^{60}Co | < 81 | < 0.036 |
| | | | |
| | ^{137}Cs | < 60 | < 0.033 |
| | | | |
| | ^{75}Se | 0.19 ± 0.06 | - |
| | | | |
| | ^{54}Mn | - | 0.11 ± 0.02 |
| | | | |
| | ^{56}Co | - | 0.08 ± 0.02 |
| | | | |
| | ^{57}Co | | 0.20 ± 0.09 |
| | | | |
| | ^{58}Co | - | 0.22 ± 0.04 |
| | | | |
| | ^{65}Zn | - | 5.2 ± 0.6 |

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| | ^{137}Cs | < 60 | < 0.033 |
| | ^{75}Se | 0.19 ± 0.06 | - |
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| | ^{57}Co | | 0.20 ± 0.09 |
| | ^{58}Co | - | 0.22 ± 0.04 |
| | ^{65}Zn | - | 5.2 ± 0.6 |



Cosmogenic nuclides

| Isotope | Decay mode | Q value, keV | Half-life, days | Activity (mBq/kg) in | |
|------------------|------------|-----------------|-----------------|----------------------|----------------------|
| | | | | “Natural” materials | “Enriched” materials |
| ⁵⁴ Mn | EC β- | 1377.1 697.1 | 312.3 | < 0.3 | 0.11(2) |
| ⁵⁶ Co | EC | 4566.0 | 77.27 | 0.3(2) | 0.08(2) |
| ⁵⁷ Co | EC | 836.0 | 271.79 | - | 0.20(9) |
| ⁵⁸ Co | EC β- | 2307.4 381.6 | 70.82 | 0.4(2) | 0.22(4) |
| ⁶⁵ Zn | EC | 1351.9 | 244.26 | 7.6(1.0) | 5.2(6) |
| ⁷⁵ Se | EC | 863.6 | 119.8 | 8.2(7) | 0.19(6) |

Less activated due to using raw material with known history, land transportation & storage underground

Cosmogenic nuclides

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Should be activated by **factor 2 less**
(according to depleting in isotopic mixture)
... and even more
because of land transportation & storage underground

Isotopic composition of Zinc

| Isotope | Recommended value | Zn grains |
|------------------|-------------------|-----------|
| ^{64}Zn | 48.6% | 47.25% |
| ^{66}Zn | 27.9% | 28.27% |
| ^{67}Zn | 4.1% | 4.24% |
| ^{68}Zn | 18.8% | 19.54% |
| ^{70}Zn | 0.6% | 0.70% |

Deviation is about 1%

Isotopic composition of Selenium

| Isotope | Recommended value | Se grains |
|------------------|-------------------|-----------|
| ^{74}Se | 0.87% | 0.73% |
| ^{76}Se | 9.36% | 8.78% |
| ^{77}Se | 7.63% | 7.35% |
| ^{78}Se | 23.78% | 23.08% |
| ^{80}Se | 49.61% | 50.53% |
| ^{82}Se | 8.73% | 9.52% |

Deviation is about 1%

Isotopic composition of enriched Selenium

| Isotope | Recommended value | ^{82}Se grains (conv. 38) |
|------------------|-------------------|------------------------------------|
| ^{74}Se | 0.87% | < 0.01% |
| ^{76}Se | 9.36% | < 0.01% |
| ^{77}Se | 7.63% | < 0.01% |
| ^{78}Se | 23.78% | < 0.01% |
| ^{80}Se | 49.61% | 4.46% |
| ^{82}Se | 8.73% | 95.52% |

Current situation

15 kg of HP Zn
6 kg of ^{82}Se

are stored deep underground @LNGS
and ready to production of ZnSe charge

A few days are required for 1 kg of Zn^{82}Se charge synthesis
(3 synthesis cycles/day + annealing in H_2 flow within 2 days)

Almost **one year** is required for whole amount of Zn^{82}Se crystal production
(1 furnace \times 3 crystals/month \times 12 months = 36 Zn^{82}Se crystals)

Summary (1)

1. Chemical and Radiopurity of HP Zn & ^{82}Se metals comply with all requirements for high quality low background ZnSe crystal production
2. R&D for natural ZnSe crystal production should be finished late January, 2015
3. First enriched Zn ^{82}Se crystal is planned to be grown late February, 2015
4. Cosmogenic activation of detector materials currently became an dominant component of internal background, and should be minimized in any possible way (*by using materials with well known history, by storage deep underground, by land transportation in shielded container, by minimization of production time, by depletion Zn metal on ^{64}Zn isotope, etc.*)
5. Small deviation in isotopic composition of Zn and Se metal allows to determine the source of raw materials and control the whole ZnSe crystal production chain on every step

nevertheless...

What we have?...

“natural” Zn – 749.2 g
HP Zn – 10080 g



“natural” Se – 500.0 g
 ^{82}Se – 2500.5 g



| Isotope | i.a. in “natural” samples, % | i.a. in “enriched” samples, % | Decay mode | $Q_{\beta\beta}$, keV |
|------------------|------------------------------|-------------------------------|-----------------------------|------------------------|
| ^{64}Zn | 47.25(5) | 47.25(5) | $\epsilon\beta^+/2\epsilon$ | 1095.7(0.7) |
| ^{70}Zn | 0.7017(6) | 0.7017(6) | $2\beta^-$ | 998.5(2.2) |
| ^{74}Se | 0. 7255(17) | < 0.01 | $\epsilon\beta^+/2\epsilon$ | 1209.169(49) |
| ^{80}Se | 50.53(9) | 4.46 | $2\beta^-$ | 133.9 |
| ^{82}Se | 9.532(11) | 95.52 | $2\beta^-$ | 2995.8 |

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HP Zn – 10080 g



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| ^{70}Zn | | | $2\beta^-$ | 998.5(2.2) |
| ^{74}Se | 0. 7255(17) | | $\epsilon\beta^+/2\epsilon$ | 1209.169(49) |
| ^{80}Se | | | $2\beta^-$ | 133.9 |
| ^{82}Se | 9.532(11) | 95.52 | $2\beta^-$ | 2995.8 |

2EC/ $\varepsilon\beta^+$ decay of ^{64}Zn isotope

| Transition | Decay channel | Level of daughter nucleus | E_γ , keV | Experimental limit $T_{1/2}$, yr at 68% C.L. | | |
|---|---------------------------------|---------------------------|------------------|---|---------------------------|--|
| | | | | “natural” Zn | HP Zn | Previous results |
| $^{64}\text{Zn} \rightarrow ^{64}\text{Ni}$ | $\varepsilon\beta^+(0\nu+2\nu)$ | g.s. | 511 | $\geq 2.7 \times 10^{21}$ | $\geq 3.2 \times 10^{22}$ | $\geq 9.4 \times 10^{20}$ for 2ν [1] $\geq 8.5 \times 10^{20}$ for 0ν [1] |
| | 2K0ν | g.s. | 1079.0 | $\geq 2.2 \times 10^{21}$ | $\geq 1.3 \times 10^{22}$ | $\geq 3.2 \times 10^{20}$ [1] |
| | KL0ν | g.s. | 1086.4 | $\geq 2.2 \times 10^{21}$ | $\geq 1.3 \times 10^{22}$ | $\geq 3.2 \times 10^{20}$ [1] |
| | 2L0ν | g.s. | 1093.7 | $\geq 2.2 \times 10^{21}$ | $\geq 1.3 \times 10^{22}$ | $\geq 3.2 \times 10^{20}$ [1] |

[1] P. Belli et al., arXiv:1110.3923v1 [nucl-ex] 18 Oct 2011.

2 β (0v+ 2v) decay of ^{82}Se to excited levels

| Transition | Decay channel | Level of daughter nucleus | Best experimental limit $T_{1/2}, \text{y}$ at 68(90)% C.L. | | |
|---|---------------|---------------------------|---|--------------------------------|--|
| | | | “natural” Se | ^{82}Se | Previous results |
| $^{82}\text{Se} \rightarrow {}^{82}\text{Kr}$ | 2 β - | 776.52 | $\geq 3.1 \times 10^{20}$ | $\geq 8.7(6.0) \times 10^{21}$ | $\geq 2.8 \times 10^{21}$ [1] $\geq 1.4 \times 10^{21}$ [2] |
| | | 1474.9 | $\geq 7.5 \times 10^{20}$ | $\geq 4.8(3.3) \times 10^{21}$ | $\geq 1.6 \times 10^{21}$ [2] |
| | | 2171.7 | $\geq 6.2 \times 10^{20}$ | | $\geq 3.0 \times 10^{21}$ [2] |

[1] Arnold *et al.*, NP A 636 (1998) 209.

[2] J. Suhonen, *et al.*, Z. Phys. A 358 (1997) 297.

Summary (2)

The low background measurements performed by HP Ge detector with “natural” and “enriched” HP Zn & ^{82}Se samples were used to set the best limits on different modes of 2β decay of ^{64}Zn and ^{82}Se isotopes to excited level of daughter nuclei at the level $\sim 10^{20}\text{-}10^{22}$ y
(first experimental results of LUCIFER project)