Risultati recenti e prospettive sullo studio del doppio decadimento beta con cristalli scintillatoti (DAMA set-up)

103º Congresso Nazionale della Società Italiana di Fisica

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Barabash A.S., Belli P., Bernabei R., Boiko R.S., Brudanin V.B., Cappella F., Caracciolo V., Cerulli R., Chernyak D.M., Danevich F.A., Incicchitti A., Kasperovych D.V., Kobychev V.V., Konovalov S.I., Laubenstein M., Mokina V.M., Poda D.V., Polischuk O.G., Shlegel V.N., Tretyak V.I., Umatov V.I., Zarytskyy M.M.

DAMA Experimental Activities

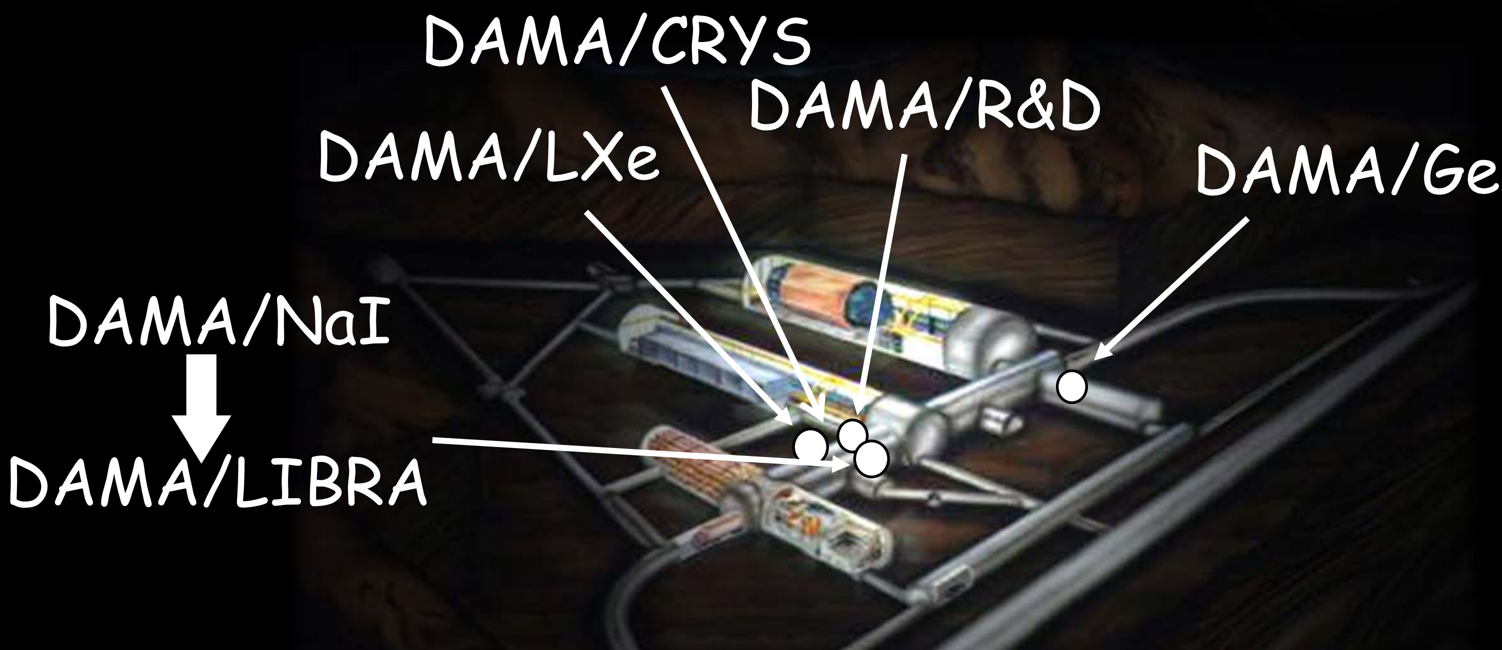
DAMA Collaboration (spokesperson: prof. R. Bernabei):

Roma2, Roma1, LNGS-INFN, IHEP/Beijing

+ by-products and small scale expts.: INR-Kiev and others

+ neutron meas.: ENEA-Frascati e ENEA-Casaccia

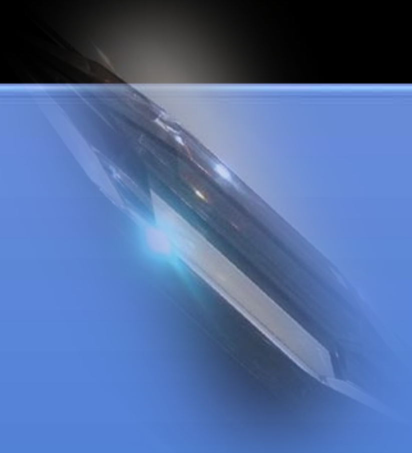
+ in some studies on ββ decays (DST-MAE project): IIT Kharagpur/Ropar, India



DAMA: an observatory for rare processes @LNGS

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

Strengths of ββ experiments by ULB crystal scintillators

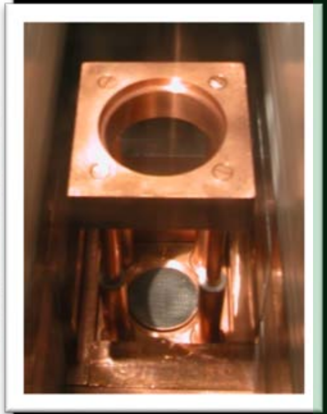


- Well known technology
- High duty cycle
- Large mass possible
- Enrichment possible in many cases
- “Ecological clean” set-up; no safety problems
- Cheaper than other considered technique
- Relatively small underground space needed
- High radiopurity by selections, chem./phys. purifications, protocols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

Esempi di isotopi utili per l'investigazione di modi di decadimento doppio beta impiegando cristalli scintillatori con la tecnica della sorgente attiva

Isotops	**Nat. Ab. (%)**	**Q (keV)**	**Decay Mode**	**Scintillator**
^{64}Zn	8.63	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	$2\beta^+$, $\varepsilon\beta^+, 2\varepsilon$	$^{106}CdWO_4$
^{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	$\varepsilon\beta^+$	$SrCl_2$, $SrI_2(Eu)$

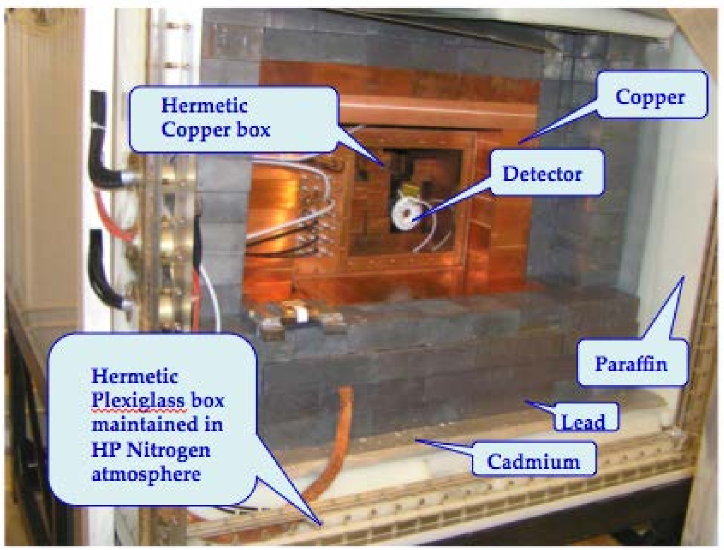
DAMA/Ge and LNGS STELLA facility - Some main previous results



Comunicazione di: F. Cappella

- RDs on highly radiopure NaI(Tl) set-up
- several RDs on low background PMTs
- qualification of many materials
- meas. on $Li_6Eu(BO_3)_3$ (NIMA572(2007)734)
- ββ decay in ^{100}Mo with the 4π low-bckg HPGe facility of LNGS (NPA846(2010)143)
- search for ^{7}Li solar axions (NPA806(2008)388, PLB711(2012)41)
- meas. with a Li_2MoO_4 (NIMA607(2009)573)
- ββ decay of ^{136}Ce and ^{138}Ce (NPA824(2009)101)
- $CdWO_4$ and $ZnWO_4$ radiopurity studies (NIMA626-627(2011)31, NIMA615(2010)301)
- First observation of α decay of ^{190}Pt to the first excited level (137.2 keV) of ^{186}Os (PRC83(2011)034603)
- ββ decay in ^{190}Pt and ^{198}Pt (EPJA47(2011)91)
- ββ decay of ^{156}Dy, ^{158}Dy (NPA859(2011)126)
- Contaminations of SrI_2(Eu) (NIMA670(2012)10)
- Radioactive contamination of ^{7}LiI(Eu) (NIMA704(2013)40)
- ββ decay of ^{96}Ru and ^{104}Ru (EPJA42(2009)171, PRC87(2013)034607)
- First search for rare decays of Os (EPJA49(2013)24)
- Search for double beta decay of ^{136}Ce and ^{138}Ce (Nucl.Phys. A930 (2014) 195-208)
- Double beta decay in ^{112}Sn and ^{124}Sn (NIMA797 (2015) 130-137)

DAMA R&D - Some main previous results:



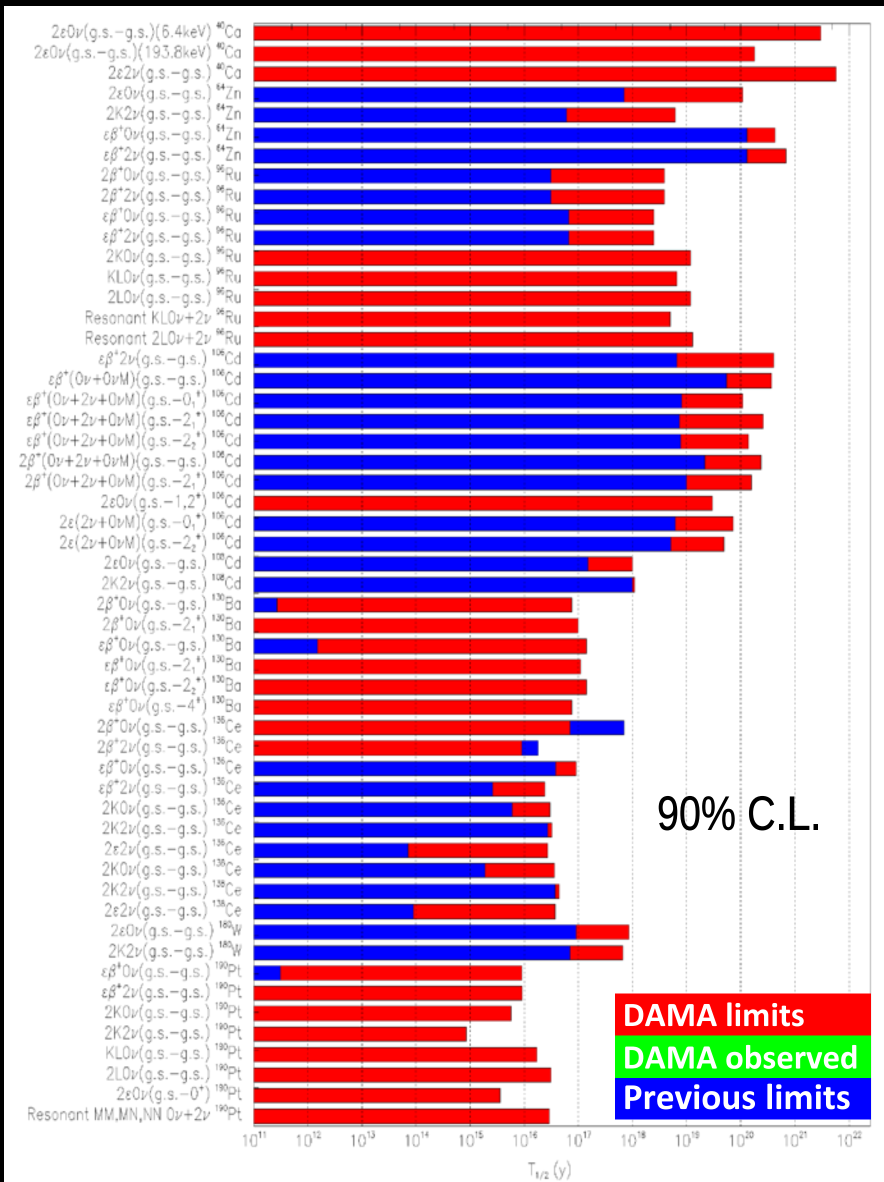
Materiale	^{238}U (ppb)	^{232}Th (ppb)	^{nat}K (ppm)
Cu	< 0.5	< 1	< 0.6
Pb boliden	< 8	< 0.03	< 0.06
Pb boliden2	< 3.6	< 0.027	< 0.06
Polish Pb	< 7.4	< 0.042	< 0.03
Polietilene	< 0.3	< 0.7	< 2
Plexiglass	< 0.64	< 27.2	< 3.3

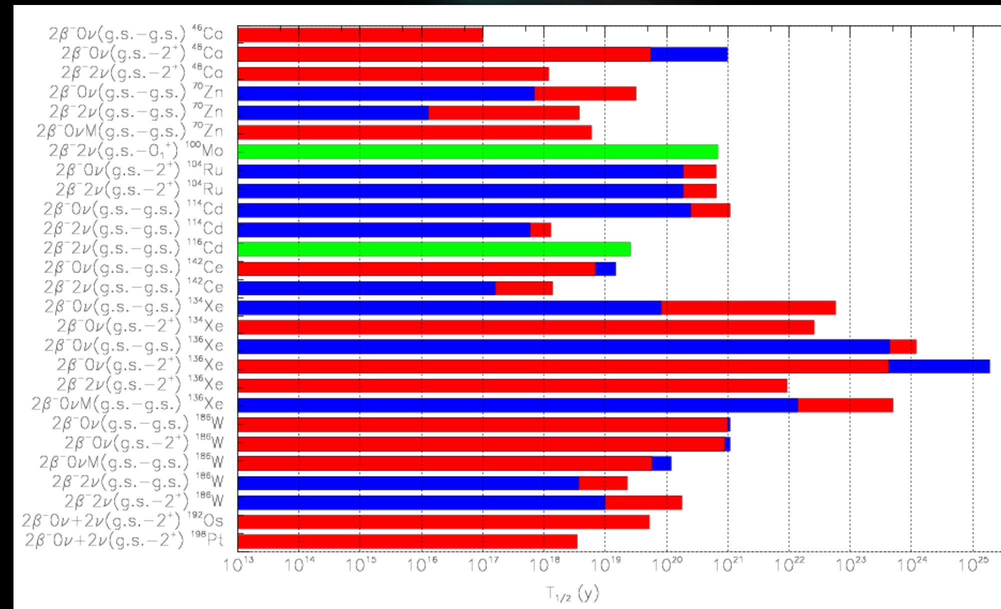
- scintillators developments: radio-purification, enrichment, optical features, etc.
- exploiting the potentiality of the low background scintillation technique to investigate rare processes with high sensitivity
- realization of pilot experiments

CaF_2(Eu), CeF_3, BaF_2, $CdWO_4$, $^{106}CdWO_4$, $^{116}CdWO_4$, $ZnWO_4$, $LaCl_3$(Ce), $LiEu(BO_3)_3$, LiF(W), $CeCl_3$, Li_2MoO_4, SrI_2(Eu), etc.

AP7(1997)73, N.Cim.A110(1997)189, NPB563(1999)97, AP10(1999)115, NPA705(2002)29, NIMA498(2003)352, NIMA525(2004)535, NIMA555(2005)270, UJP51(2006)1037, NPA789(2007)15, PRC76(2007)064603, PLB658(2008)193, EPJA36(2008)167, NPA824(2009)101, NPA826(2009)256, JPG:NPP38(2011)115107, JPG: NPP38(2011)015103, JINST6(2011)P08011, PRC85(2012)044610, EPJC73(2013)2276, EPJA50(2014)134, PS90(2015)085301

Summary of searches for ββ decay modes in various isotopes (partial list)





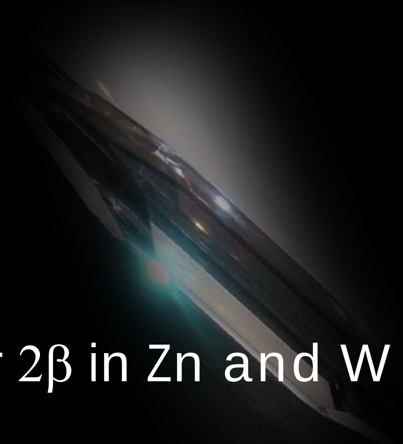
ARMONIA: New observation of $2\nu2\beta^-$ $^{100}Mo \rightarrow {}^{100}Ru$ ($g.s. \rightarrow 0_1^+$) decay NPA846 (2010)143

AURORA: New observation of $2\nu2\beta^-$ ^{116}Cd decay J.Phys.:Conf.Ser.718(2016)062009

- 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, ^{184}Os, ^{156}Dy, ^{158}Dy, …).
- First searches for resonant $0\nu2\varepsilon$ decays in some isotopes

In the following present

STATUS and PERSPECTIVES on:



- ✓ $ZnWO_4$ crystal scintillators to search for 2β in Zn and W isotopes
- ✓ $^{106}CdWO_4$/$^{116}CdWO_4$ crystal scintillators to search for 2β in ^{106}Cd/^{116}Cd
- ✓ Improvement of crystal radio-purity by recrystallization technique
- ✓ $SrI_2(Eu)$ crystal scintillator for low-level counting experiments

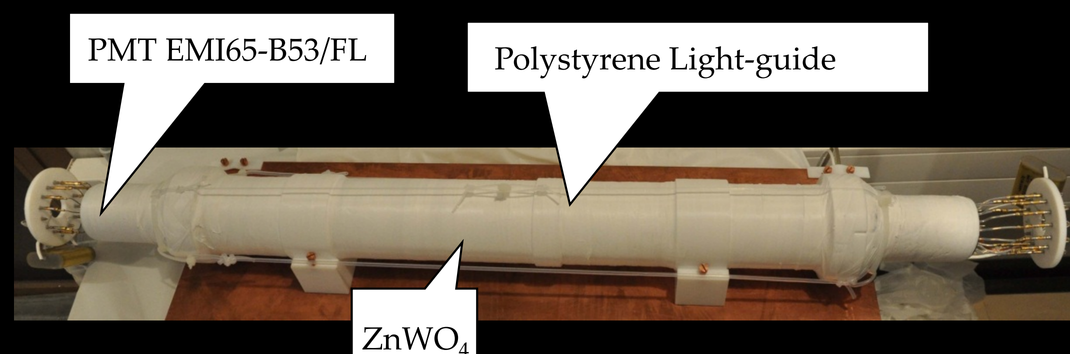
$ZnWO_4$ crystal scintillators

- Development of low background $ZnWO_4$ crystal scintillators with large volume and high scintillation properties is important to investigate double beta decay modes in Zn and W isotopes with source=detector approach

Transition	Energy release ($Q_{\beta\beta}$) (keV) [26]	Isotopic abundance (%) [27]	Decay channels	Number of mother nuclei in 100 g of $ZnWO_4$ crystal
$^{64}Zn \rightarrow {}^{64}Ni$	1095.7(0.7)	49.17(75)	2ε, $\varepsilon\beta^+$	9.45×10^{22}
$^{70}Zn \rightarrow {}^{70}Ge$	998.5(2.2)	0.61(10)	$2\beta^-$	1.17×10^{21}
$^{180}W \rightarrow {}^{180}Hf$	144(4)	0.12(1)	2ε	2.31×10^{20}
$^{186}W \rightarrow {}^{186}Os$	489.9(1.4)	28.43(19)	$2\beta^-$	5.47×10^{22}

PLB658(2008)193
NPA826(2009)256
NIMA626-627(2011)31
JP38(2011)115107
EPJC73 1(2013) 2276
PS90 8(2015)085301
NIMA833 (2016) 77-81

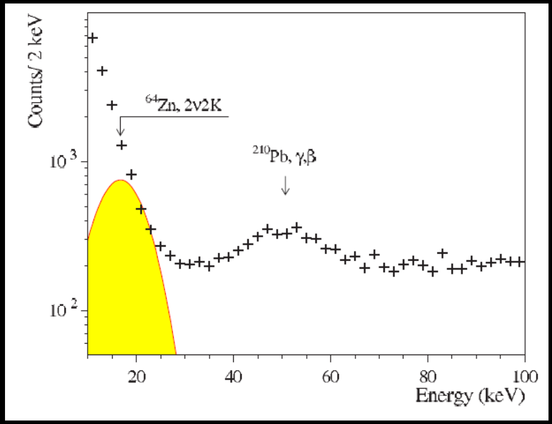
- Various detectors with mass 0.1-0.7 kg realized by exploiting different materials and techniques
- Inside a cavity (filled up with high-pure silicon oil) ϕ 47 x 59 mm in central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length.



Final results on the present stage of investigation of ββ decay modes in Zn and W isotopes with low background $ZnWO_4$

JPGNPP 38 (2011) 115107

Total exposure = 0.529 kg x y



Improved (up to 2 orders of magnitude) $T_{1/2}$ limits on ββ decay modes of ^{64}Zn, ^{70}Zn, ^{180}W and ^{186}W:

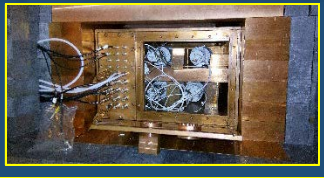
now at level of $10^{18} – 10^{21}$ yr

→ up to now only 5 nuclides (^{40}Ca, ^{78}Kr, ^{112}Sn, ^{120}Te and ^{106}Cd) over 34 candidates to 2ε, $\varepsilon\beta^+$, $2\beta^+$ processes have been studied at similar level of sensitivity in direct search experiments

1. A possible positive hint of the (2n+0n)$\varepsilon\beta^+$ decay in ^{64}Zn with $T_{1/2} = (1.1 \pm 0.9) \times 10^{19}$ yr [Bikit et al., Appl. Radiat. Isot. 46(1995)455] excluded
2. the 0ν2β capture in ^{180}W is of particular interest because of possible resonant process;
3. the rare α decay of the ^{180}W with $T_{1/2} = (1.3^{+0.6}_{-0.5}) \times 10^{18}$ yr observed and new limit on the $T_{1/2}$ of the α transition of the ^{183}W to the metastable level $1/2^-$ at 375 keV of ^{179}Hf has been set: $T_{1/2} = 6.7 \times 10^{20}$ yr.

$ZnWO_4$ - Work in Progress…

- New 4 crystal scintillators in the DAMA/R&D in data taking:
  - radioactive contamination and scintillation performances
  - study of double beta decay modes in Zn and W isotopes



Development of Detectors with Anisotropic Response
For Dark Matter Search in Directionality Approach are in progress

- Cryostat for low temperature measurement with scintillation, testing in progress
- detectors realized
- Lowering the energy threshold
- Measurements of anisotropy at low energy with Neutron Generator in progress at Casaccia ENEA lab
- Development of electronics

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

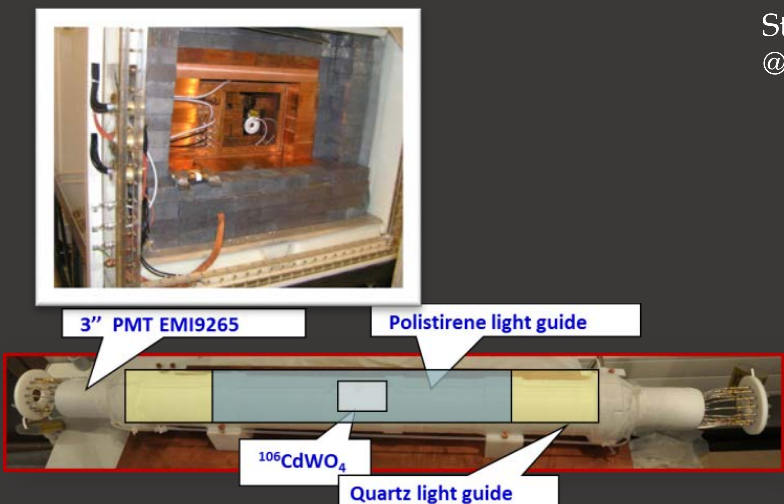
The used $^{106}CdWO_4$ crystal scintillator

NIMA615(2010)301

- Samples of cadmium were purified by vacuum distillation (Institute of Physics and Technology, Kharkiv) and the Cadmium tungstate compounds were synthesized from solutions
- Crystal boule was grown by the low-thermal-gradient Czochralski technique (NIIC Novosibirsk) (initial powder 265 g)
- Crystal scintillator (**216 g** mass), **66.4%** enrichment in ^{106}Cd (**2.66×10^{23}** nuclei of ^{106}Cd) measured by thermal ionisation mass-spectrometry ⇒ 2nd enriched $CdWO_4$ crystal ever produced

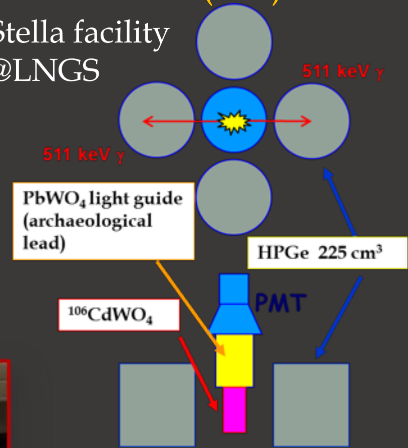
1st exp: single crystal in DAMA/R&D

PRC85(2012)044610

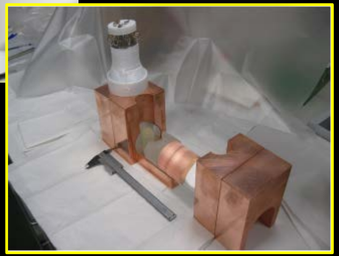


2nd exp: coincidence with 4 HP-Ge

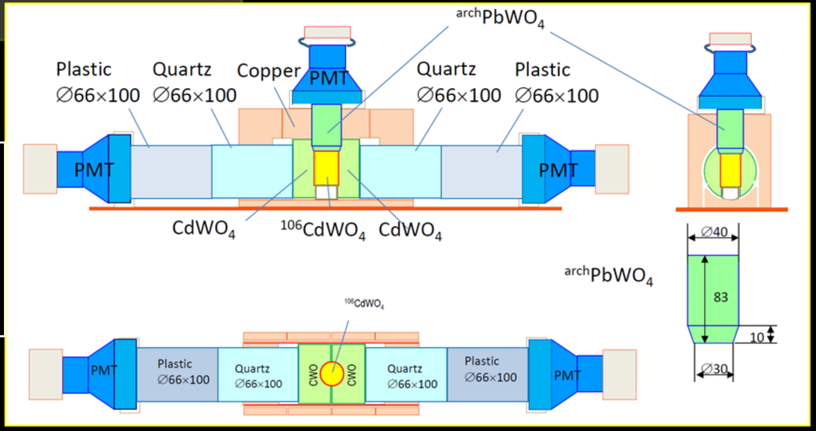
PRC93(2016)045502



3th exp

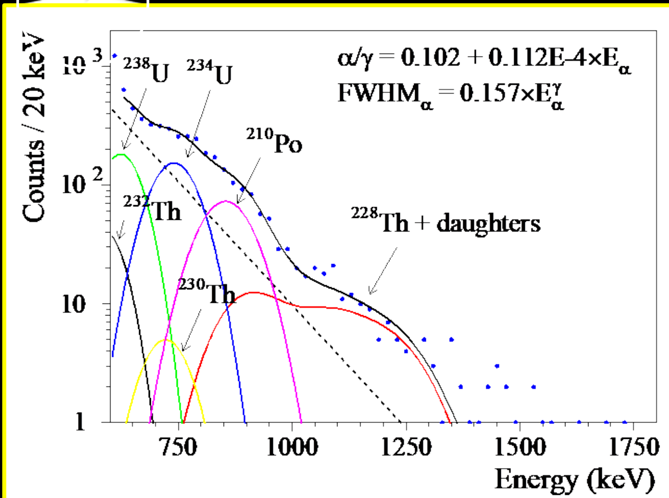


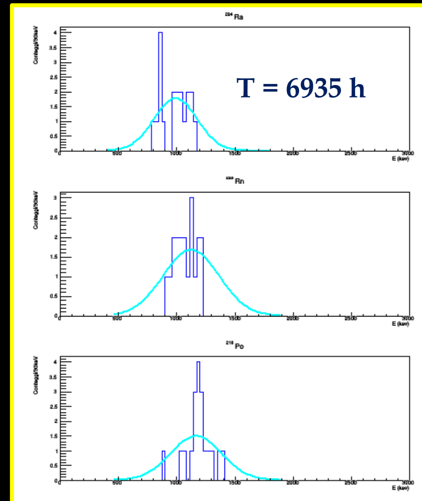
$^{106}CdWO_4$ in (anti)coincidence with two large $CdWO_4$ scintillators mounted in DAMA/CRYS set-up @ LNGS



New ^{106}CdWO4 experiment in DAMA/CRYS set-up

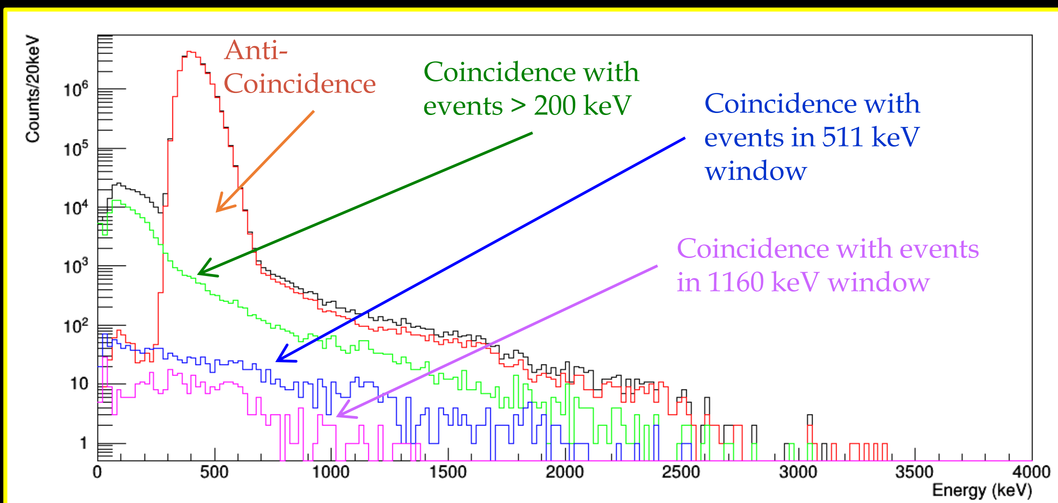
Preliminary





The arrival time, the energy and the pulse shape of each event were used to select the fast decay chain in the ^{228}Th sub-chain of the ^{232}Th family in $^{106}CdWO_4$ crystal:

^{224}Ra (Q = 5.789 MeV, $T_{1/2}$ = 3.66 d)
↓
^{220}Rn (Q = 6.405 MeV, $T_{1/2}$ = 55.6 s)
↓
^{216}Po (Q = 6.906 MeV, $T_{1/2}$ = 0.145 s)
↓
^{212}Pb



The energy spectra accumulated over **6935 h** by the $^{106}CdWO_4$ detector:

- in anticoincidence with the $^{nat}CdWO_4$ detectors
- in coincidence with event(s) in at least one of the $^{nat}CdWO_4$ detectors with energy:
  - E > 200 keV
  - E in energy window around 511 keV
  - E in energy window around 1160 keV

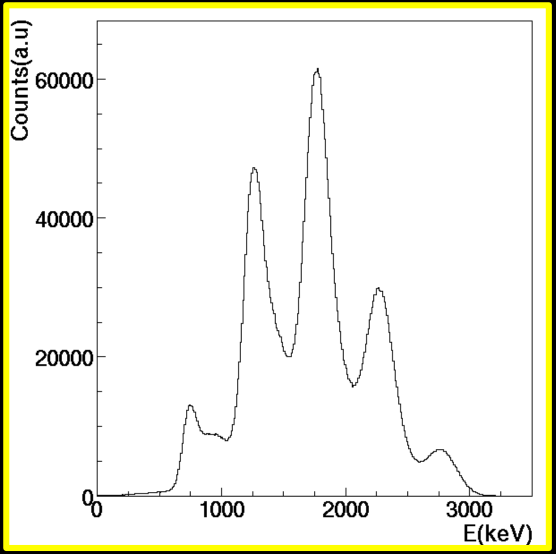
⇒ Activity of ^{228}Th: **5(1) μBq/kg**
⇒ Estimation of α/γ light ratio
⇒ Estimation of α energy resolution

Chain	Nuclide	a (mBq/kg)
^{232}Th	^{232}Th	<0.07
	^{228}Th+subch.	<0.02
^{238}U	^{238}U	<0.6
	^{234}Th	<0.6
	^{230}Th	<0.4
	^{210}Po	<0.2

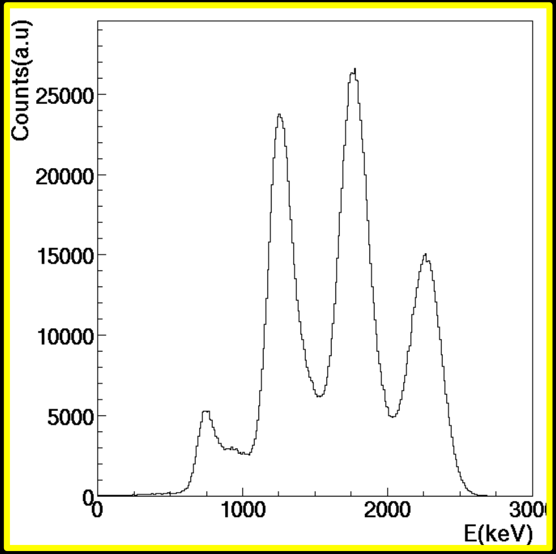
Estimation of sensitivity

Expected signal for ^{106}Cd $0\nu 2\beta^+(0^+ \rightarrow 0^+)$:

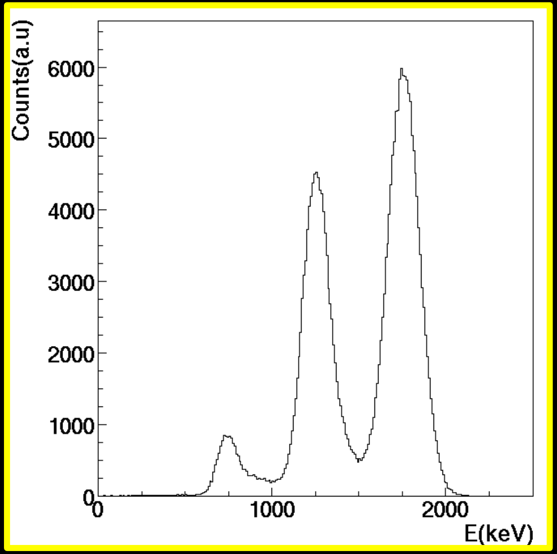
Spectrum of $^{106}CdWO_4$ detector



Spectrum of $^{106}CdWO_4$ detector when one of the two $CdWO_4$ detectors detects γ of 511 keV ($\pm 2\sigma$)



Spectrum of $^{106}CdWO_4$ detector when both the $CdWO_4$ detectors detect γ of 511 keV ($\pm 2\sigma$)



Sensitivity after 1yr in the hypothesis of about 30 background counts in [0.-3.] MeV:

$0\nu\ \varepsilon\beta^+$ (g.s.): $T_{1/2} \approx 5 \times 10^{21}$ yr

$2\nu\ 2\beta^+$ (g.s.): $T_{1/2} \approx 2 \times 10^{21}$ yr

In the region of theoretical predictions: $T_{1/2} \sim 10^{20} – 10^{22}$ yr

Note that, up to now, 2ν mode of the $2\beta^+$ processes has not been detected unambiguously: there are only indications for ^{130}Ba and ^{78}Kr

The AURORA experiment in the DAMA/R&D set-up: Investigation of 2β decay of ^{116}Cd with enriched $^{116}CdWO_4$ crystal scintillators

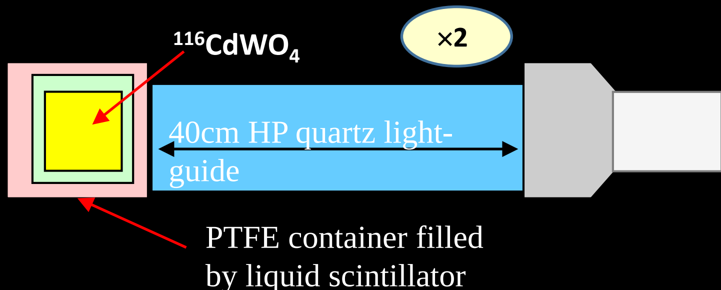
^{116}Cd: one of the best isotope for 0ν2β decay search:

- $Q_{\beta\beta}$ = 2813.44(13) keV
- δ = 7.49(18)%
- possible high isotopic enrichment
- promising theoretical calculation

$^{116}CdWO_4$ crystal scintillators

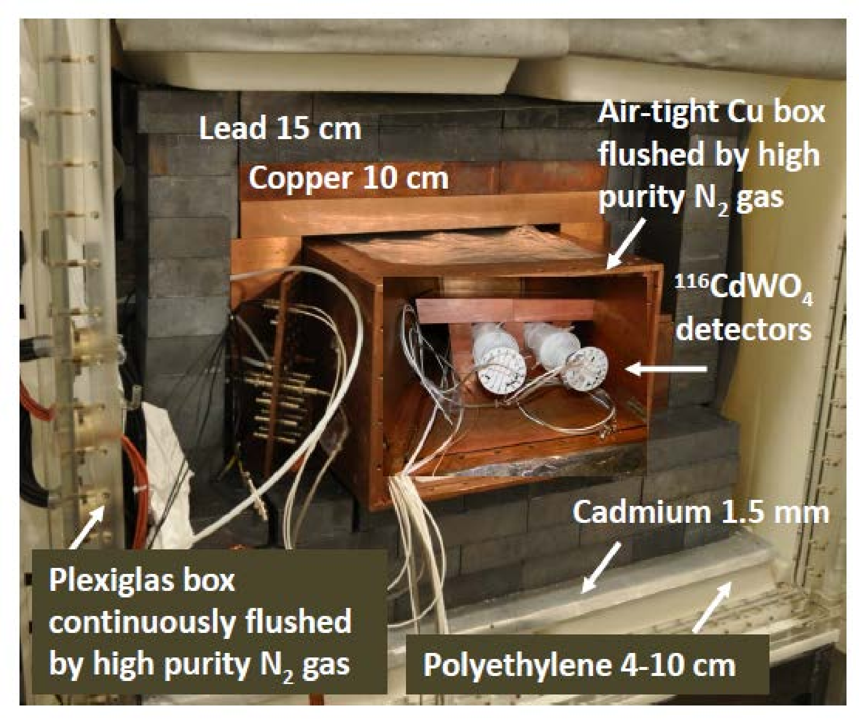
Grown by the low-thermal-gradient Czochralski technique after deep purification of ^{116}Cd and W;
+ annealing to improve the optical transmission curve

- ✓ Good optical and scintillation properties
- ✓ $^{116}CdWO_4$ crystals enriched at 82%
- ✓ Active source approach (high detection efficiency)
- ✓ Low levels of internal contamination in (U, Th, K)
- ✓ α/β discrimination capability



Two enriched $^{116}CdWO_4$ crystal scintillators
(total mass: 1.162 kg, ^{116}Cd @ 82%)

- ✓ Started in 2011
- ✓ Upgrade - March 2014
- ✓ Total live time since 2014: 25037 h
- ✓ Background level at 2.7-2.9 MeV: 0.1 counts/keV/kg/yr

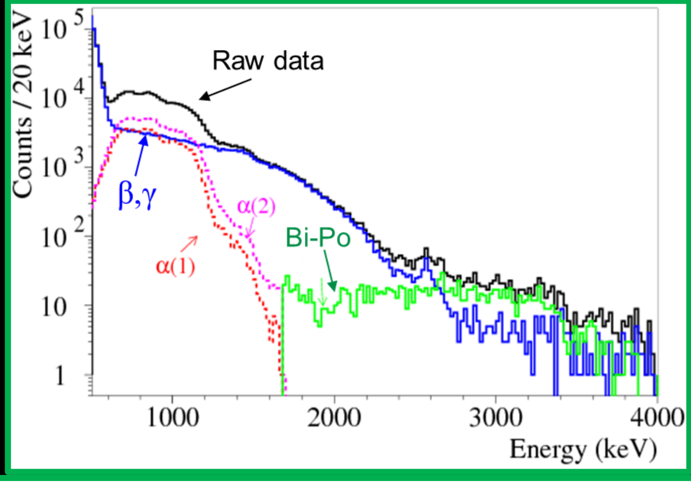


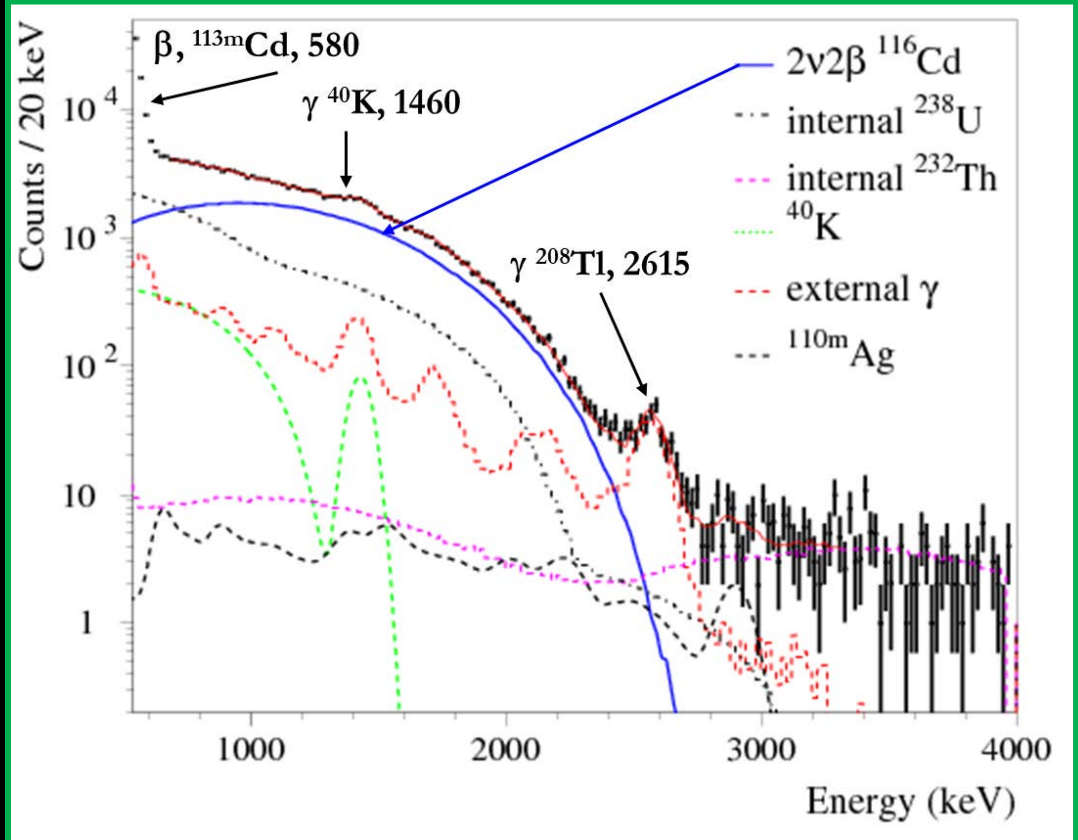
AURORA Experiment: Background Identification

Radioactive contaminations of $^{116}CdWO_4$ crystal scintillators

Chain	Nuclide	Activity mBq/kg
^{232}Th	^{232}Th	0.61(2)
	^{228}Th	0.022(3)
^{238}U	^{238}U	0.59(7)
	^{234}Th	0.64(7)
	^{230}Th	0.11(2)
	^{226}Ra	≤ 0.01
	^{210}Pb	0.6(1)
	^{40}K	0.20(1)
	^{110m}Ag	<0.06

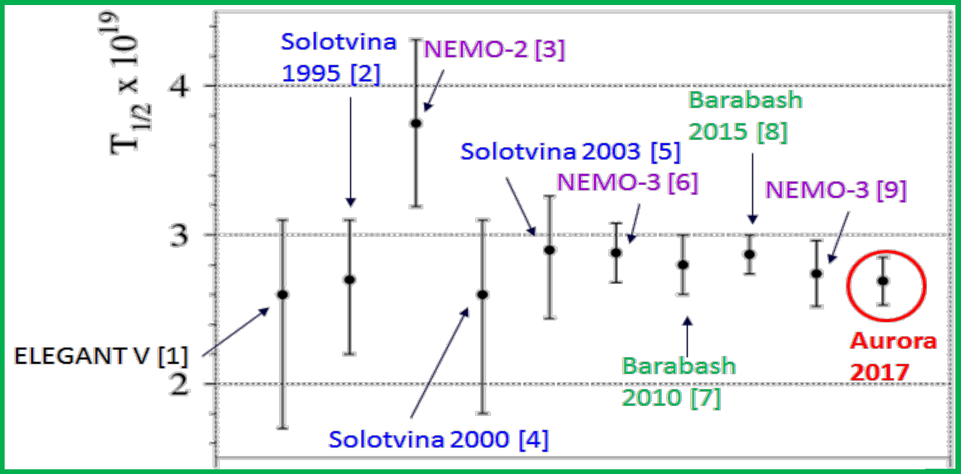
Total α activity = 2.27 mBq/kg





AURORA Experiment: Result for $2\nu2\beta$ Decay of ^{116}Cd & $T_{1/2}$ Limit on $0\nu2\beta$ Decay of ^{116}Cd

$T_{1/2}$ = [2.69 ± 0.02(stat.) ± 0.14(syst.)] × 10^{19} yr (the most accurate value up to date)



[1] J. Phys. Soc. Japan 64(1995)339
[2] Phys. Lett. B 344(1995)72
[3] Z. Phys. C 72(1996)239
[4] PRC 62(2000)045501
[5] PRC 68(2003)035501
[6] AIP Conf. Proc. 1572(2013)110
[7] PRC 81(2010)035501
[8] NPA 935(2015)52
[9] PRD 95(2017)012007

Fit in 2.5–3.2 MeV: -3.7 ± 10.6 counts

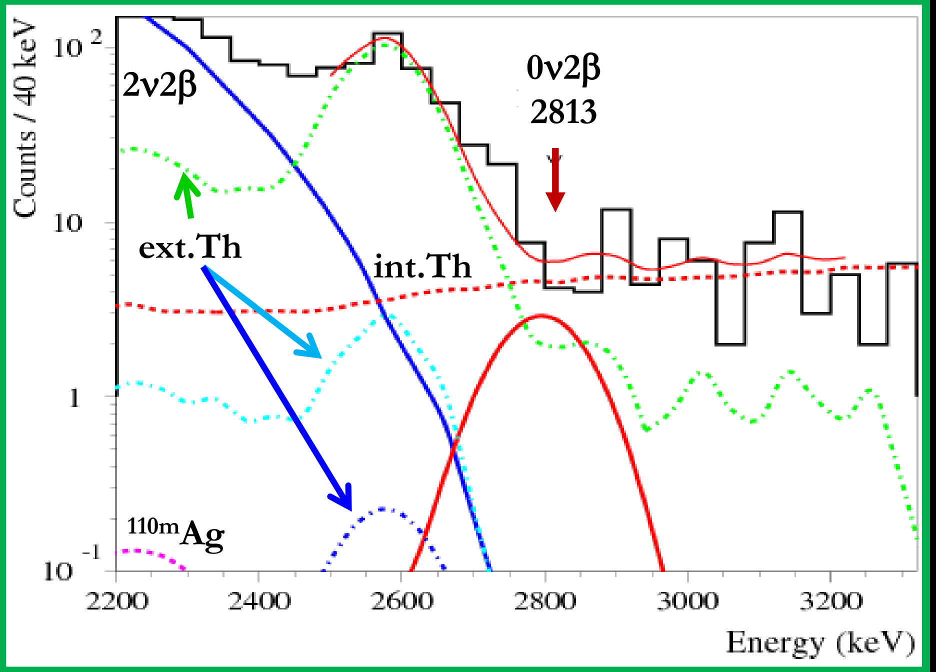
$T_{1/2} > 2.4 \times 10^{23}$ yr @ 90% C.L.

Effective Majorana neutrino mass:

$\langle m_\nu \rangle < 1.1 - 1.6$ eV [1-4]

+

New improved limits on $T_{1/2}$ for $0\nu2\beta$ decay to excited levels of ^{116}Sn in the range:
(3.6–6.3)×10^{22} yr



[1] T.R. Rodryguez et al., Phys.Rev.Lett. 105(2010)252503
[2] F. Simkovic et al., Phys.Rev.C 87 (2013)045501
[3] J. Hyvarinen et al., Phys.Rev.C 91 (2015)024613
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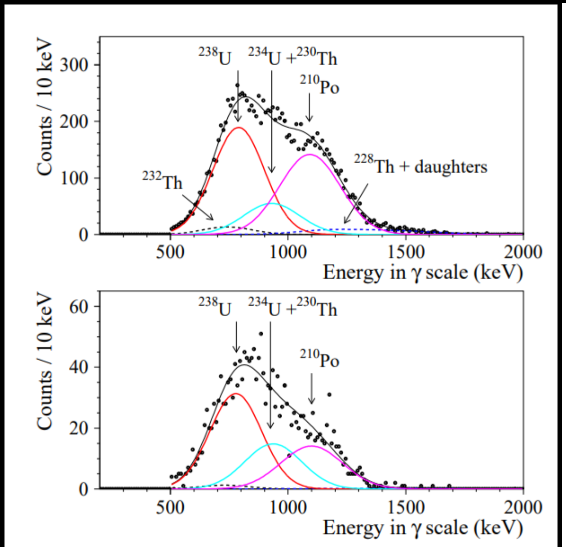
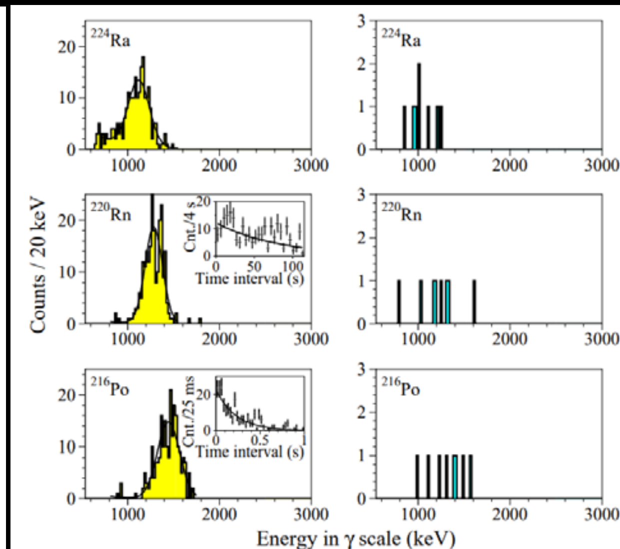
Improvement of radiopurity of ^{116}CdWO4 by recrystallization

A.S. Barabash et al., Nucl. Instr. Meth. A 833(2016)77

Re-crystallized by the low-thermal-gradient Czochralski technique in a platinum crucible





Crystal n.3 used (326 g mass)

60% of initial mass after re-crystallization

Side surface made opaque by grinding paper to improve light collection

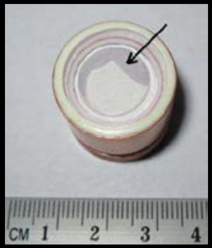
Radioactive contamination of the samples (before an after recrystallization) measured in the DAMA/CRYS setup @ LNGS

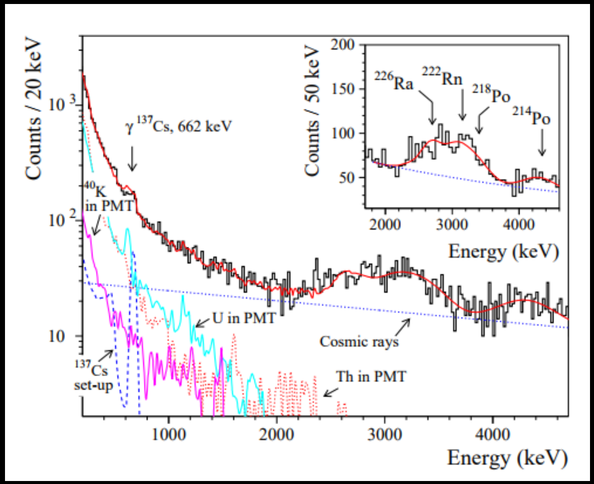
Chain	Nuclide (sub-chain)	Activity (mBq/kg)	
		Before recrystallization	After recrystallization
^{232}Th	^{232}Th	0.13(7)	0.03(2)
	^{228}Th	0.10(1)	0.010(3)
^{238}U	^{238}U	1.8(2)	0.8(2)
	^{226}Ra	≤0.1	≤0.015
	^{234}U+^{230}Th	0.6(2)	0.4(1)
	^{210}Po	1.6(2)	0.4(1)
Total α		4.44(4)	1.62(4)

- ^{228}Th reduced by a factor ~10 ⇒ 0.01 mBq/kg
- α activity reduced by a factor ~3 ⇒ 1.6 mBq/kg

main background component for ^{116}Cd $0\nu2\beta$ decay

Feasibility Study for Low-Level Counting Experiment by $SrI_2(Eu)$ Crystal Scintillator





Features in literature

Density: $(4.5 – 4.6)$ g/cm^3

Melting point: 515 ◦C

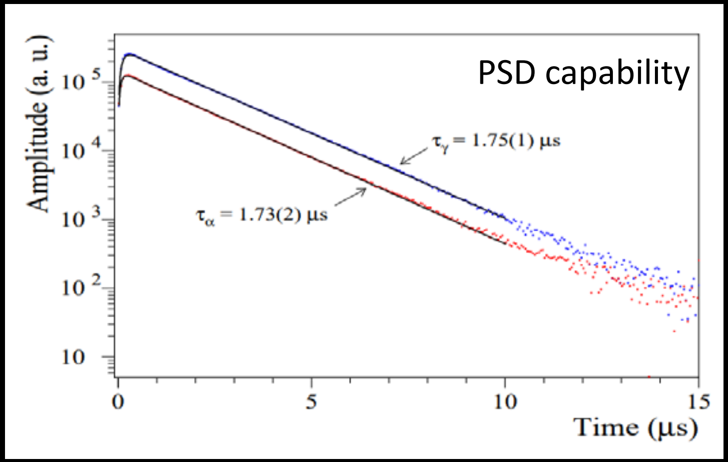
Index of refraction: 1.85

Wavelength of emission maximum: 429 – 436 nm

Light yield: (68 – 120) photons/keV

Energy resolution (FWHM) for 662 keV γ: (2.6 – 3.7)%

Scintillation decay time at 300 K: (0.6 – 2.4) μs



- A single crystal of SrI_2 doped by 1.2% of Eu was grown in a quartz ampoule using the vertical Stockbarger method
- Studied radioactive contamination and scintillation property
- Applicability of $SrI_2(Eu)$ to the search for ββ decay of ^{84}Sr was demonstrated for the first time.
- New improved half-life limits were set on 2ε and $\varepsilon\beta^+$ decay in ^{84}Sr at level of $T_{1/2} \sim 10^{15} – 10^{16}$ yr.

Process of decay	Decay mode	Level of daughter nucleus (keV)	E_γ (keV)	η	$T_{1/2}$ (yr) Present work	[35, 36]
$\varepsilon\beta^+$	0ν	g.s.	511	7.2%	$> 6.9 \times 10^{15}$	$> 7.3 \times 10^{13}$
$\varepsilon\beta^+$	2ν	g.s.	511	7.2%	$> 6.9 \times 10^{15}$	–
$2K$	0ν	g.s.	1754 – 1762	4.0%	$> 6.0 \times 10^{16}$	–
KL	0ν	g.s.	1767 – 1775	3.9%	$> 1.9 \times 10^{16}$	–
$2L$	0ν	g.s.	1779 – 1788	3.9%	$> 5.9 \times 10^{16}$	–
2ε	0ν	2^+ 881.6	881.6	5.0%	$> 2.6 \times 10^{16}$	–
2ε	2ν	2^+ 881.6	881.6	5.8%	$> 3.1 \times 10^{16}$	–

Work in Progress....

- Radio-purity improvements
- Studying different crystal growing technique
- Growing large volume $SrI_2(Eu)$ crystal scintillator to study its radioactive contamination and scintillation performances

Conclusions

- ✓ Many and competitive results have been obtained in the search for $\beta\beta$ decay by the DAMA experimental set-ups at LNGS
- ✓ Continue efforts to develop new/improved crystal scintillators for low bckg physics
- ✓ Experiments on 2β decay of Zn and W isotopes running/under-improvement
- ✓ Experiments for development of crystal scintillators with anisotropic response for nuclear recoil in keVee region are in progress
- ✓ Experiments on 2β decay of ^{106}Cd and ^{116}Cd running/under-improvement
- ✓ Search for 2β processes in ^{116}Cd with $^{116}CdWO_4$ (enriched to 82%) scintillation detectors (1.16 kg) just concluded in the DAMA/R&D set-up:
  - $T_{1/2}(2\nu2\beta)$ = [2.69 ± 0.02(stat.) ± 0.14(syst.)] × 10^{19} yr (the most accurate value up to date)
  - $T_{1/2}(0\nu2\beta) \geq 2.4 \times 10^{23}$ yr → $\langle m\nu \rangle < (1.1 - 1.6)$ eV (the best limit)
  - Internal ^{228}Th (main bkgd) can be strongly reduced by re-crystallization
- ✓ Studies for the Improvement of crystal scintillators' radiopurity are in progress
- ✓ Feasibility Study for Low-Level Counting Experiment by SrI_2(Eu)Crystal Scintillator