DAMA Collaboration & INR-Kyiv

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# **Results and perspectives in ββ decay experiments by the DAMA-Kyiv Collaboration with HPGe**

103° Congresso SIF, September 2017



# DAMA: an observatory for rare processes @LNGS

## DAMA/CRYS

DAMA/R&D DAMA/Ge

DAMA/LXe

DAMA/NaI

DAMA/LIBRA



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### Collaboration

#### Roma Tor Vergata, Roma La Sapienza, LNGS, IHEP/Beijing



+ by-products and small scale experiments (MoU): INR-Kyiv



+ in some studies on  $\beta\beta$  decays (DST-MAE projects, inter-univ. Agreem.): IIT Ropar/Kharagpur, India





#### + in some activites collaborators from

Ukraine	Kyiv National Taras Shevchenko University National Science Center Kharkiv Instit. of Physics and Technology; Institute for Scintillation Materials, Ukraine
Russia	Russian Chemistry-Technological University of D.I.Mendeleev Moscow Joint Institute for Nuclear Research, Dubna; Joint stock company NeoChem, Moscow Nikolaev Inst. of Inorganic Chemistry, Novosibirsk; Institute of Theoretical and Experimental Physics, Moscow
Australia	Department of Applied Physics, Curtin University, Perth
Finland	Dept. of Physics, University of Jyvaskyla, Jyvaskyla

# Summary of searches for $\beta\beta$ decay modes in various isotopes (partial list)



# DAMA/Ge and LNGS STELLA facility

### Ge detectors used by DAMA in previous searches:

#### DAMA/Ge (GeBer)

- 244 cm<sup>3</sup> n-type HPGe detector
- Thin Carbon window: 0.76 mm thickness

### GeCris

- 465 cm<sup>3</sup> p-type HPGe detector
- Thin Cu window: 1 mm thickness

#### GeMulti

- Four 225 cm<sup>3</sup> p-type HPGe detectors mounted in one cryostat with a well in the center
- Thin Al window: 1.3 mm thickness

#### GeBEGe

- Broad Energy Ge detector (especially designed for low energy γ spectrometry)
- Thin Cu window: 1.5 mm thickness

### **DAMA** results

- Search for ββ decays of many candidate isotopes (next slide)
- Search for <sup>7</sup>Li solar axions (NPA806(2008)388, PLB711(2012)41)
- First observation of  $\alpha$  decay of <sup>190</sup>Pt to the first excited level of <sup>186</sup>Os (PRC83(2011)034603)
- Qualification of many materials: e.g. CdWO<sub>4</sub>, ZnWO<sub>4</sub>(NIMA626-7(2011)31, NIMA615(2010)301), Li<sub>6</sub>Eu(BO<sub>3</sub>)<sub>3</sub> (NIMA572(2007)734), Li<sub>2</sub>MoO<sub>4</sub> (NIMA607(2009)573), SrI<sub>2</sub>(Eu) (NIMA670(2012)10), <sup>7</sup>LiI(Eu) (NIMA704(2013)40)

Typical shield from environmental radioactivity

- 5-10 cm of OFHC copper
- 5 cm of low activity lead (< 3 Bq/kg of <sup>210</sup>Pb)
- 15-25 cm of lead
- 10 cm of borated polyethylene (GeBer)
- Air-tight PMMA box flushed with HP nitrogen



### First or improved results for 2β decays of many isotopes

<sup>136</sup>Ce  $Q_{\beta\beta}$ =2378.55 keV; 2ε, εβ<sup>+</sup>, 2β<sup>+</sup>; <sup>138</sup>Ce  $Q_{\beta\beta}$ =691 keV; 2ε

- > CeO<sub>2</sub> sample (627 g) in GeCris detector (2299 h)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>17</sup>-10<sup>19</sup> yr [Eur. Phys. J. A 53 (2017) 172]
- > CeO<sub>2</sub> sample (732 g) in GeCris detector (1900 h)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>17</sup>-10<sup>18</sup> yr [Nucl. Phys. A 930 (2014) 195]
- > CeCl<sub>3</sub> crystal (6.9 g) in DAMA/Ge detec. (1280 h)  $\Rightarrow$  T<sub>1/2</sub> limits: (1÷6)10<sup>15</sup> yr [Nucl. Phys. A 824 (2009) 101]

#### <sup>106</sup>Cd $Q_{\beta\beta}$ =2775.39 keV; 2 $\epsilon$ (res 0 $\nu$ ), $\epsilon\beta^+$ , 2 $\beta^+$ [Phys. Rev. C 93 (2016) 045502]

>  $^{106}$ CdWO<sub>4</sub> crystal scintillator (216 g) in GeMulti (13085 h)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>20</sup>-10<sup>21</sup> yr

<sup>96</sup>Ru Q<sub>ββ</sub>=2714.51 keV; 2ε (res 0ν), εβ<sup>+</sup>, 2β<sup>+</sup>, <sup>104</sup>Ru Q<sub>ββ</sub>=1301.2 keV; 2β<sup>-</sup>

- > Purified Ru samples in GeMulti det. (0.56kg×yr)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>20</sup>-10<sup>21</sup> yr [Phys. Rev. C 87 (2013) 034607]
- > Ru sample (473 g) in GeCrys detector (158 h)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>18</sup>-10<sup>19</sup> yr [Eur. Phys. J. A 42 (2009) 171]

<sup>184</sup>Os  $Q_{\beta\beta}$ =1453.7 keV; 2 $\epsilon$  (res 0 $\nu$ ),  $\epsilon\beta^+$ ; <sup>192</sup>Os  $Q_{\beta\beta}$ =412.4 keV; 2 $\beta^-$  [Eur. Phys. J. A 49 (2013) 24]

> Os sample (173 g) in GeCris detector (2741 h)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>16</sup>-10<sup>17</sup> yr for <sup>184</sup>Os and 10<sup>19</sup> yr for <sup>192</sup>Os

<sup>190</sup>Pt  $Q_{\beta\beta}$ =1383 keV; 2 $\epsilon$  (res 0 $\nu$ ),  $\epsilon\beta^+$ ; <sup>198</sup>Pt  $Q_{\beta\beta}$ =1049 keV; 2 $\beta^-$  [Eur. Phys. J. A 47 (2011) 91]

> Pt sample (42.5 g) in GeCris detector (1815 h)  $\Rightarrow$  T<sub>1/2</sub> limits: 10<sup>14</sup>-10<sup>16</sup> yr for <sup>190</sup>Pt and 10<sup>18</sup> yr for <sup>198</sup>Pt

<sup>156</sup>Dy Q<sub>ββ</sub>=2005.95 keV; 2ε, εβ<sup>+</sup>; <sup>158</sup>Dy Q<sub>ββ</sub>=282.7 keV; 2ε [Nucl. Phys. A 859 (2011) 126] > Dy<sub>2</sub>O<sub>3</sub> sample (322 g) in DAMA/Ge det. (2512 h) ⇒ T<sub>1/2</sub> limits: 10<sup>14</sup>-10<sup>16</sup> yr

<sup>100</sup>Mo  $Q_{\beta\beta}$ =3035 keV; 2 $\beta^{-1}$ 

[Nucl. Phys. A 846 (2010) 143]

<sup>100</sup>MoO<sub>3</sub> sample (1199 g) enriched in <sup>100</sup>Mo at 99.5% in GeMulti detector ⇒ observation of <sup>100</sup>Mo→<sup>100</sup>Ru(0<sub>1</sub><sup>+</sup>) decay:  $T_{1/2} = 6.9^{+1,0}_{-0,8}(stat) \pm 0.7(syst) \times 10^{20}$  yr

The best experimental sensitivities in the field for  $2\beta$  decays with positron emission

# Armonia

NPA846(2010)143



#### (meAsuReMent of twO-NeutrIno $\beta\beta$ decAy of <sup>100</sup>Mo to 0<sup>+</sup><sub>1</sub> level of <sup>100</sup>Ru )

In addition to the transition to the g.s., the  $2\beta 2\nu$  decay of  $^{100}Mo$  was registered also for the transition to the first excited  $0^{+}_{1}$  level of  $^{100}Ru$ 

- If O<sup>+</sup><sub>1</sub> excited level of <sup>100</sup>Ru (E=1130 keV) populated
- $\Rightarrow$  two  $\gamma$  quanta (591 keV + 540 keV) emitted in cascade



<sup>100</sup>MoO<sub>3</sub> sample (mass =1199 g) enriched in <sup>100</sup>Mo at 99.5% installed in GeMulti setup



T <sub>1/2</sub> measured in several experiments:	$T_{1/2}, 10^{20}  ext{ yr}$	Year [Ref.]
Frejus UL (4800 m w.e.), HP Ge 100 cm <sup>3</sup> , 994 g of <sup>100</sup> Mo (99.5%), 2298 h, only 1-d spectrum;	> 12	1992 [19]
Soudan mine (2090 m w.e.), HP Ge 114 cm <sup>3</sup> , 956 g of <sup>100</sup> Mo (98.5%), 9970 h, 1-d spectrum;	$6.1^{+1.8}_{-1.1}$	1995 [11] <sup>a</sup>
Modane UL (4800 m w.e.), 4 HP Ge detectors (100, 120, 380, 400 cm <sup>3</sup> ), 17 different <sup>100</sup> Mo samples (107–1005 g, 95.1–99.3%, 142–1599 h), sum of 1-d spectra;	$9.3^{+2.8}_{-1.7}$	1999 [14]
Modane UL (4800 m w.e.), NEMO-3 detector, 6914 g of <sup>100</sup> Mo foils in 12 sectors (95.1–98.9%), 8024 h, individual energies of $\gamma$ and $e^-$ , tracks for $e^-$ ;	$5.7^{+1.5}_{-1.2}$	2007 [15]
Ground level (10 m w.e.), 2 HP Ge detectors (300 cm <sup>3</sup> ) in coincidence, 1050 g of <sup>100</sup> Mo (98.4%), 21720 h, coincidence spectrum;	$5.5^{+1.2}_{-0.9}$	2009 [16] <sup>b</sup>
Gran Sasso UL (3600 m w.e.), 4 HP Ge detectors (225 cm <sup>3</sup> each) in coincidence, 1199 g of <sup>100</sup> MoO <sub>3</sub> (99.5%), 18120 h, coincidence and 1-d spectra.	$6.9^{+1.2}_{-1.1}$	This work

Aim of the experiment: remeasurement of the Mo sample used before in the Frejus exp. (not in agreement with other results)

# **Armonia Results**

#### NPA846(2010)143



### 2-dim energy spectrum analysis

Double coincidences when fixing the energy of one of the Ge detectors

Eight events detected (red)

$$T_{1/2} = 6.8^{+3.7}_{-1.8}$$
(stat.) × 10<sup>20</sup> yr

in agreement with the half life derived in 1-d analysis

### 1-dimensional energy spectrum analysis

Both peaks at 540 keV and 591 keV expected for  $2\beta 2\nu$ decay  ${}^{100}\text{Mo} \rightarrow {}^{100}\text{Ru}(0_1^+)$  are observed in the data collected with  ${}^{100}\text{Mo}O_3$ 

In the background spectrum they are absent

Fit of peak @ 539.5 keV: E=539.4 $\pm$ 0.2 keV; S<sub>540</sub> = 319 $\pm$ 56 events Fit of peak @ 590.8 keV: E= 590.9 $\pm$ 0.2 keV; S<sub>591</sub> = 278 $\pm$ 53 events

$$T_{1/2} = 6.9^{+1.0}_{-0.8}$$
(stat.)  $\pm 0.7$ (syst.)  $\times 10^{20}$  yr.

Most of systematic unc. due to calculation of the efficiencies



# Search for ββ decay in <sup>106</sup>Cd with <sup>106</sup>CdWO<sub>4</sub> scintillator in coincidence with 4 HPGe (GeMulti)

### <sup>106</sup>Cd, a promising isotope:

- ✓ One of the six isotopes candidate for  $2\beta^+$  decay
- ✓  $\delta = (1.25 \pm 0.06)\% \Rightarrow$  possible enrichment up to 100%
- ✓  $Q_{2\beta}$ = (2775.39±0.10) keV  $\Rightarrow$  2β<sup>+</sup>, εβ<sup>+</sup>, 2ε modes possible
- ✓ Possible resonant 2ε0v captures to excited level of <sup>106</sup>Pd
- ✓ Theoretical  $T_{1/2}$  favorable for some 2v modes (10<sup>20</sup> 10<sup>22</sup> yr)





### <sup>106</sup>CdWO<sub>4</sub> crystal scintillator:

- ✓ Mass: 216 g, 66.4% enrichment in <sup>106</sup>Cd
- ✓ Good scintillation properties
- ✓ Active source approach (high detection efficiency)
- ✓ Low levels of internal contamination in (U, Th K)
- ✓  $\alpha/\beta$  discrimination capability

### PbWO<sub>4</sub> light-guide (Ø40 × 83 mm)

Reduce PMT background (archael. lead: A(<sup>210</sup>Pb)<0.3 mBq/kg)

## <sup>106</sup>CdWO<sub>4</sub> crystal scintillator in GeMulti: Results



Energy spectrum of <sup>106</sup>CdWO<sub>4</sub> detector in coincidence with 511 keV in HPGe (circles). Monte Carlo simulated distributions of 2β decay of <sup>106</sup>Cd excluded at 90% CL

- 1. In anticoincidence with the HPGe detectors (AC)
- 2. In coincidence with  $E_{HPGe} > 200 \text{ keV}$  (CC >200)
- 3. In coincidence with  $E_{HPGe} = 511 \text{ keV}$  (CC 511)
- 4. In coincidence with  $E_{HPGe} = 1160 \text{ keV}$  (CC 1160)



- > New limits on 2 $\epsilon$ ,  $\epsilon\beta^+$ ,  $2\beta^+$  processes on the level of  $T_{1/2} > 10^{20} 10^{21}$  yr
- The half-life limit on the εβ<sup>+</sup>2ν decay, T<sub>1/2</sub> > 1.1×10<sup>21</sup> yr, reached the region of theoretical predictions
- ► For 2 $\epsilon$ 0v resonant captures:  $T_{1/2}$  > (8.5 × 10<sup>20</sup> 1.4 × 10<sup>21</sup>) yr

## New <sup>106</sup>CdWO<sub>4</sub> experiment in DAMA/Crys set-up

- New experiment with <sup>106</sup>CdWO<sub>4</sub> in (anti)coincidence with two large CdWO<sub>4</sub> scintillators mounted in DAMA/Crys set-up @ LNGS
- 2) High efficiency
- 3) Experiment in data taking since May 2016





### New limits on $2\beta^+$ decay of <sup>136</sup>Ce and <sup>138</sup>Ce with deeply purified cerium sample

2315.3

2222.7

2141.3

2128.8

2080.0

1579.0

1551.0

818.5

 $Q_{20} = 2378.55(27)$  ke



Ce purification performed by the liquid-liquid extraction method

improved 2 $\beta$  sensitivity  $\approx$  one order of magnitude

 $\Rightarrow$  thorium concentration reduced by a factor  $\approx 60$ 

No peculiarities in CeO<sub>2</sub> spectrum can be ascribed to  $2\beta$  decay of <sup>136</sup>Ce or <sup>138</sup>Ce  $\Rightarrow$  New improved half-life limits:  $T_{1/2} > 10^{17} - 10^{19} \text{ yr}$ 



<sup>136</sup><sub>56</sub>Ba

#### Eur. Phys. J. A 53 (2017) 172

	Chain	in Nuclide	Activity (mBq $kg^{-1}$ )		
			before	after 1st	after 2nd
			purification	[31] purification	[31] purification
		$^{40}\mathrm{K}$	77(28)	$\leq 9$	$\leq 4$
		$^{137}\mathrm{Cs}$	$\leq 3$	$\leq 2$	$0.4 \pm 0.2$
		$^{138}La$	1000 h	$\leq 0.7$	$\leq 0.6$
_		$^{139}\mathrm{Ce}$	-	$6\pm1$	$1.4 \pm 0.3$
		$^{152}\mathrm{Eu}$		$\leq 0.5$	$\leq 0.2$
7		$^{154}\mathrm{Eu}$		$\leq 0.9$	$\leq 0.08$
		<sup>176</sup> Lu	-	$\leq 0.5$	$0.4 \pm 0.1$
	$^{232}\mathrm{Th}$	$^{228}$ Ra	$850\pm50$	$53 \pm 3$	$30.4\pm0.7$
		$^{228}\mathrm{Th}$	$620\pm30$	$573 \pm 17$	$9.8 \pm 0.5$
	$^{235}\mathrm{U}$	$^{235}\mathrm{U}$	$38 \pm 10$	$\leq 1.8$	$\leq 0.4$
		$^{231}$ Pa		$\leq 24$	$\leq 0.4$
		$^{227}\mathrm{Ac}$		$\leq 3$	$\leq 1.4$
	$^{238}\mathrm{U}$	$^{238}\mathrm{U}$	$\leq 870$	$\leq 40$	$\leq 12$
		$^{226}$ Ra	$11\pm3$	$\leq 1.5$	$\leq 0.3$

#### NB:

Cerium purification is also motivated in the light of radiopure crystal scintillators development; In fact, Ce is used

- ✓ to develop Ce-containing crystal scintillators (e.g., CeF<sub>3</sub>, CeCl<sub>3</sub>)
- $\checkmark$  as a dopant in inorganic scintillators as Gd<sub>2</sub>SiO<sub>5</sub>(Ce), YAlO<sub>3</sub>(Ce), LaBr<sub>3</sub>(Ce)

# Running and future experiments

0+ 8×10<sup>18</sup> y (1-) 2.68 h

150Nd

150Pm

150Sm

- Experiment running since February 2015 with deeply purified  $Nd_2O_3$  sample (2381) g) in GeMulti detector to investigate 2 $\beta$  decay of <sup>150</sup>Nd to excited levels of <sup>150</sup>Sm:
  - $\Rightarrow$  Background rate in the region of expected peaks (334.0 keV and 406.5 keV) ≈ 2 counts/keV/d
  - $\Rightarrow$  Expected T<sub>1/2</sub> sensitivity after 500 days of measurements: 1.3×10<sup>20</sup> yr (90%CL)



		Q <sub>2β</sub> =	3368			
Improvement of Nd <sub>2</sub> O <sub>3</sub> radioactive contamination						
Contamination	Before [1]	Present				
<sup>40</sup> K	46	< 4				
<sup>214</sup> Bi ( <sup>226</sup> Ra)	1.1	<0.4				
<sup>228</sup> Ac ( <sup>228</sup> Ra)	0.9	<0.4				



- New experiment to search for 2 $\beta$  of osmium (and  $\alpha$  decay of osmium) to excited level of daughter nuclei) in progress with BEGe detector:
  - $\Rightarrow$  Detection efficiency significantly improved by cutting the osmium roads into thin (0.8-1 mm) plates and by using the **BEGe detector**
- Purification of Er, Yb, and Sm is in progress for experiments to search for resonant  $2\varepsilon 0v$  processes in these nuclei



# Conclusions

Many and competitive results have been obtained by the DAMA-Kyiv collab. in the search for  $\beta\beta$  decays with HPGe detectors @ the STELLA facility of LNGS:

- ✓ First or improved limits on the half-lives of double beta decays of <sup>96</sup>Ru, <sup>104</sup>Ru, <sup>106</sup>Cd, <sup>112</sup>Sn, <sup>124</sup>Sn, <sup>136</sup>Ce, <sup>138</sup>Ce, <sup>156</sup>Dy, <sup>158</sup>Dy, <sup>184</sup>Os, <sup>192</sup>Os, <sup>190</sup>Pt and <sup>198</sup>Pt
- The best experimental sensitivities in the field for  $2\beta$  decays with positron emission (useful to distinguish the mechanism of neutrinoless  $2\beta$  decay)
- $\checkmark$  Possible resonant 2:0v processes investigated in several candidate isotopes
- New observation of the 2β2v decay of <sup>100</sup>Mo to the first excited 0<sup>+</sup><sub>1</sub> level of <sup>100</sup>Ru with the coincidence technique in the ARMONIA experiment
- ✓ New and competitive limits on  $2\epsilon$ ,  $\epsilon\beta^+$ ,  $2\beta^+$  processes of  ${}^{106}Cd$  with a  ${}^{106}CdWO_4$ detector in coincidence with 4 HPGe detetors ( $T_{1/2} > 10^{20}-10^{21}$  yr, reached the region of theoretical predictions for the  $\epsilon\beta^+2\nu$  decay)