Roma2, Roma1, LNGS, IHEP/Beijing

+ in some small scale expts and by-products results: INR-Kiev
 + in the framework of MAE activity: IIT-Kharagpur
 + neutron measurements: ENEA- Frascati



13° Lomonosov Conf. Moscow 23 - 29 Aug. 2007

Search for rare processes at Gran Sasso

DAMA/LXe DAMA/R&D

DAMA/NaI

DAMA/LIBRA (see R. Bernabei talk 29/8)

F. Nozzoli INFN & Univ. Roma Tor Vergata low bckg DAMA/Ge for sampling meas.

_meas. with ¹⁰⁰Mo

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

DAMA/NaI(Tl) ~100 kg

Results on DM particles:

• PSD

- PLB389(1996)757
- Investigation on diurnal effect
 N.Cim.A112(1999)1541
- Exotic Dark Matter search PRL83(1999)4918
- Annual Modulation Signature

PLB424(1998)195, PLB450(1999)448, PRD61(1999)023512, PLB480(2000)23, EPJC18(2000)283, PLB509(2001)197, EPJ C23 (2002)61, PRD66(2002)043503, Riv.N.Cim.26 n.1 (2003)1-73, IJMPD13(2004)2127, IJMPA21(2006)1445, EPJC47(2006)263 + IJMPA22(2007)3155 + others in progress

total exposure collected in 7 annual cycles

Results on rare processes:

- Possible Pauli exclusion principle violation
- CNC processes
- Electron stability and non-paulian transitions in Iodine atoms (by L-shell)

Riv.N.Cim.26 n. 1(2003)1-73, IJMPD13(2004)2127

- Search for solar axions
- Exotic Matter search
- Search for superdense nuclear matter
- Search for heavy clusters decays

PLB408(1997)439 PRC60(1999)065501

PLB460(1999)235 PLB515(2001)6 EPJdirect C14(2002)1 EPJA23(2005)7 EPJA24(2005)51



Performances: N.Cim.A112(1999)545-575, EPJC18(2000)283,

data taking completed on July 2002

107731 kg×d

DAMA/LXe

Unique low-background 6.5 kg LXe set-up operating as pure scintillator filled by Kr-free Xenon enriched either in ¹²⁹Xe (99.5%) or in ¹³⁶Xe (68.8%)

DARK MATTER investigations ...

- Limits on recoils investigating the DMp-¹²⁹Xe elastic scattering by PSD PLB436(1998)379
- Limits on DMp-¹²⁹Xe inelastic scattering PLB387(1996)222, NJP2(2000)15.1
- Neutron calibrations PLB436(1998)379, EPJdirectC11(2001)1

... and other rare processes

- Electron decay into invisible channels AstroP5(1996)217
- Nuclear level excitation of ¹²⁹Xe during CNC processes PLB465(1999)315
- N, NN decay into invisible channels in ¹²⁹Xe PLB493(2000)12
- Electron decay $e^- \rightarrow v_e \gamma \text{ PRD61(2000)117301}$
- 2β decay in ¹³⁴Xe PLB527(2002)182
- 2β in ¹³⁴Xe & ¹³⁶Xe PLB546(2002)23
- CNC decay $^{136}Xe \rightarrow ^{136}Cs$ Beyond the Desert (2003) 365
- N, NN, NNN decay into invisible channels in ¹³⁶Xe EPJA27 s01 (2006)35

DAMA/R&D

- Particle Dark Matter search with CaF₂(Eu) NPB563(1999)97, AP7(1997)73
- 2 β decay in ¹³⁶Ce and in ¹⁴²Ce II N.Cim.A110(1997)189
- 2EC2v ⁴⁰Ca decay AstroP7(1999)73
- 2 β decay in ⁴⁶Ca and in ⁴⁰Ca NPB563(1999)97
- $2\beta^+$ decay in ¹⁰⁶Cd AstroP10(1999)115



- 2β and β decay in ⁴⁸Ca NPA705(2002)29
 - 2EC2ν in ¹³⁶Ce, in ¹³⁸Ce and α decay in ¹⁴²Ce NIMA498(2003)352
 - $2\beta^+0\nu$ and $EC\beta^+0\nu$ decay in ^{130}Ba
 - NIMA525(2004)535
 - Cluster decay in LaCl₃(Ce) NIMA555(2005)270
 - CNC decay 139 La $\rightarrow ^{139}$ Ce UkrJP51(2006)1037
 - Rare α decay of ¹⁵¹Eu Nucl. Phys. A.798(2007)15
 - 2β processes in ⁶⁴Zn Prep. ROM2F/2007/13 +several other results under analysis + data

taking in progress + many other meas. scheduled



• measurements with 100 Mo sample investigating some double beta decay mode in progress in the 4π lowbackground HP Ge facility of LNGS (in the volume Nucl.Phys. and Atomic Energy,ed. INR-Kiev (2007), 479)

+ Many other meas. already scheduled



- stand-by during all the period of the problems related to the use of "liquids" underground because of Borexino accident.
- restarted on May 2004, running till December 2004; then stopped again waiting for the necessary restoring of circulating water in the underground lab.
- Full revision + new chiller -> restart of data taking in 2007

DAMA/Ge & LNGS Ge facility

low background Ge detector operative in the Ge facility of LNGS mainly devoted to measurements and selections for:

- RD-I, RD-II and now RD-III on highly radiopure NaI(Tl) set-ups;
- several RDs on low background PMTs;
- qualification of many materials
- measurements with a $Li_6Eu(BO_3)_3$ crystal (NIMA572(2007)734)



DAMA results on ββ decay



Experimental limits on T_{1/2} obtained by DAMA (red) and by previous experiments (blue)

(all the limits are at 90% C.L. except for $2\beta^+0\nu$ in ¹³⁶Ce and $2\beta^-0\nu$ in ¹⁴²Ce - 68% C.L.)

Now in progress measurement on $2\beta 2\nu$ decay in ¹⁰⁰Mo to the first excited 0⁺₁ level of ¹⁰⁰Ru by using ~1 kg of Molybdenum enriched in ¹⁰⁰Mo at 99.5% inserted in a 4 HP Ge detector array

Search for a decay of natural Europium

NIM A 572 (2007) 734 Nucl.Phys.A798(2007)15

Both natural Europium isotopes, ¹⁵¹Eu (natural abundance \bar{o} = 47.81(6)%) and ¹⁵³Eu (\bar{o} = 52.19(6)%) have a positive energy release respectively to a decay and, thus, they are potentially a radioactive.



The low Q_{α} for ¹⁵³Eu gives no hope for experimental observation of its decay due to the very big expected half-life.

However, for ¹⁵¹Eu estimations of half-life give values on the level of 10¹⁸ y, and such a sensitivity can be reached with current experimental techniques.



Search for α decays of Eu with a Li₆Eu(BO₃)₃ crystal

NIM A 572(2007)734

Greater content of Eu with respect to e.g. CaF₂(Eu), but poor scintillation properties and small size crystals

<u>Exp set-up</u>: Li₆Eu(BO₃)₃, 2.72 g, as external source + HP Ge detector (408 cm³) in low-background set-up @ LNG5 a

Spectra measured with the HP Ge

Radioactive contamination in $Li_{A}Eu(BO_{3})_{3}$:

Chain	Nuclide	Activity (Bq/kg)
²³² Th	²²⁸ Ac	< 0.20
	²¹² Pb	< 0.25
	²⁰⁸ Tl	< 0.13
²³⁸ U	²¹⁴ Pb	< 0.17
	²¹⁴ Bi	< 0.07
	40 K	<1.5
	⁶⁰ Co	< 0.026
	¹³⁷ Cs	< 0.081
	²⁰⁷ Bi	< 0.009
	¹⁵² Eu	= 0.949(48)
	¹⁵⁴ Eu	= 0.212(35)
Limits are given at 90%	C.L.	



¹⁵²Eu and ¹⁵⁴Eu contamination

Radioactive ¹⁵²Eu and ¹⁵⁴Eu nuclei were produced by neutron capture by ¹⁵¹Eu and ¹⁵³Eu, respectively

 $\begin{aligned} & \text{Cross section for capture of thermal neutron: } \sigma_{151} = 5900 \pm 200 \text{ b}, \quad \sigma_{153} = 312 \pm 7 \text{ b} \\ & \text{Number of nuclei: } N_{151} = 7.78 \times 10^{20}, N_{153} = 8.49 \times 10^{20} \\ & \text{Decay constant: } \lambda_{152} = \frac{\ln 2}{13.537} \text{ yr}^{-1}, \quad \lambda_{154} = \frac{\ln 2}{8.593} \text{ yr}^{-1} \end{aligned} \qquad \begin{aligned} & \text{Mass-spectrometric measurements showed that the Eu in the crystal was of normal composition: } \\ & \text{True and 52.1(3)}^{N_{153}} = \frac{1}{12} \text{ gr}^{-1}, \quad \lambda_{154} = \frac{\ln 2}{8.593} \text{ gr}^{-1} \end{aligned} \qquad \begin{aligned} & \text{Mass-spectrometric measurements showed that the Eu in the crystal was of normal composition: } \\ & \text{True and 52.1(3)}^{N_{153}} \text{Eu} \end{aligned}$ $& \Rightarrow \quad \frac{dN_d}{dt} = \phi \cdot \sigma_m \cdot N_m - \lambda_d \cdot N_d \quad \Rightarrow \quad N_d(t) = \phi \cdot \sigma_m \cdot N_m \cdot \frac{\left(1 - e^{-\lambda_d t}\right)}{\lambda_d} \quad \text{where } \begin{cases} \text{m = 151 (153)} \\ \text{d = 152 (154)} \end{cases}$ $& \frac{A_{152}}{A_{154}} = \frac{\lambda_{152} \cdot N_{152}(t)}{\lambda_{154} \cdot N_{154}(t)} = \frac{\sigma_{151} \cdot N_{151}}{\sigma_{153} \cdot N_{153}} \cdot \frac{\left(1 - e^{-\lambda_{154}t}\right)}{\left(1 - e^{-\lambda_{154}t}\right)} \end{aligned}$

 $\Rightarrow \text{ for given N}_{\text{m}} \text{ and } \sigma_{\text{m}}: \quad 11.00 < A_{152}/A_{154} < 17.33$ inconsistent with the measured ratio $A_{152}/A_{154} = 4.48 \pm 0.77$

Can be due to non-thermal neutrons flux from spontaneous fission of Th and U, and from (α,n) reactions in the monazite ores (which is a commercial source for both Eu and Th elements).

Alpha decays of Eu to Pm

NIM A 572(2007)734





 $T_{1/2}$ > 2.4 \times 10¹⁶ yr (90% C.L.)

¹⁵³Eu (g.s.) \rightarrow ¹⁴⁹Pm (g.s.) – very low Q_a=274 keV –

No evidence for 286 keV γ emitted in the β decay of ¹⁴⁹Pm (yield=3.1%, ε =11.0%)

 $T_{1/2} > 1.1 \times 10^{16} \text{ yr}$ (90% C.L.)

Search for α decays of Eu with a CaF₂(Eu) crystal Nucl. Phys. A798(2007)15

- > Low background CaF₂(Eu) scintillator, 3" diameter, 1" in length \rightarrow 370 g mass
- Pure quartz light guide (TETRASIL-B), 10 cm long to
- Low background photomultiplier (PMT) EMI9265-B53/FL.

(Scintillation crystal and light guide wrapped by PTFE reflection tape)

Eu concentration: ICP-MS (0.4 ± 0.1)%









signals recorded by a 160 MSa/s Transient Digitizer over a 3125 ns time window

DAMA/R&D set-up at LNGS

Low radioactive Cu box flushed with HP N₂

Passive shield

- 10 cm of high purity Cu
- 15 cm of low radioactive lead
- 1.5 mm of cadmium and
- 4 to 10 cm of polyethylene/paraffin

The whole shield closed inside a Plexiglas box also flushed with HP $\rm N_{2}$



Response to γ quanta and α particles

Nucl.Phys.A798(2007)15

Energy scale and resolution for γ quanta measured with ²²Na, ¹³³Ba, ¹³⁷Cs, ²²⁸Th and ²⁴¹Am sources. FWHM_{γ}(keV) = $\sqrt{3.7(4) \cdot E_{\gamma}}$ [keV]

 $1 \text{ with}_{\gamma}(\text{kev}) = \sqrt{3.7(4)} E_{\gamma}[\text{kev}]$

(e.g.: 28% @ 60 keV; 11.4% @ 356 keV and 7.5% @ 662 keV)



Energy dependence of the α/β ratio

Nucl.Phys.A798(2007)15



Non-linear behaviour near 2 MeV \Rightarrow fit by polynomial function (continuous line)

 $\alpha/\beta = 0.128(19)$ at the energy of ¹⁵¹Eu α particles

 \Rightarrow ¹⁵¹Eu α peak expected at 245(36) keV (in gamma scale)



Despite rather low discrimination, SI allows to check the nature of background

Measured energy spectrum (7426 h)

Nucl.Phys.A798(2007)15



Pulse-shape discrimination useful to identify a active nuclides in the crystal.

Background identification: Time-amplitude analysis

Arrival time and energy of each event used for the selection of fast decay chains in ²³²Th and ²³⁵U families.



Background identification: Bi-Po events

Nucl.Phys.A798(2007)15

Selection of double pulses produced by very fast decay chains (the so called Bi-Po events) depressed by a factor ≈ 110 because of the 丁 = 7426 5 used TD time window ²³²Th family 20 keV Counts / 20 keV 30 α^{212} Po (a) (b) $\alpha/\beta = 0.245(2)$ ²¹²Bi (Q_B=2.254 MeV) Counts / 75. $\rightarrow {}^{212}$ Po (Q_a=8.954 MeV, T_{1/2}=0.299 μ s) α^{214} Po 50 $\alpha/\beta = 0.221(4)$ \rightarrow ²⁰⁸Pb 25 -²²⁸Th activity = 0.124(5) mBq/kg 1000 2000 2000 3000 3000 4000 1000 4000 (in agreement with time-amplitude analysis) Energy (keV) Energy (keV) $\begin{array}{c} \text{Stimulation}\\ \text{Stimu$ Counts / 6.25 ns (c) $T_{1/2} = 0.294(12) \,\mu$ 238U family

10

1

Ó

0.5

1.5

2.5

 $\Delta t (\mu s)$

200

400

Time bin

²¹⁴Bi (Q_B=3.272 MeV) \rightarrow ²¹⁴Po (Q_a=7.833 MeV, T_{1/2}=164 μ s) \rightarrow ²¹⁰Pb

²²⁶Ra activity = 1.3(2) mBq/kg

Radioactive contamination of the $CaF_2(Eu)$

Nucl.Phys.A798(2007)15

Activities of a active nuclides in CaF2(Eu) estimated by analysis of identified a peaks

Presence in the crystal of β active isotopes (U/Th families, ${}^{40}K$, ${}^{60}Co$, ${}^{90}Sr-{}^{90}Y$, ${}^{137}Cs$, ${}^{154}Eu$) and external γ rays simulated with GEANT4



<u>A†</u>	low energy	the	background	is	caused	mainly
by	<u><u><u></u><u></u><u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u><u></u></u></u></u>	152 E	Ц			

Chain	Nuclide	Activity mBq/kg
²³² Th	²³² Th ²²⁸ Ra ²²⁸ Th	$\begin{array}{l} 0.05(1) \ \leq 0.6 \ 0.13(2) \end{array}$
$^{235}\mathrm{U}$	$^{227}\mathrm{Ac}$	0.011(7)
$^{238}\mathrm{U}$	²³⁸ U ²²⁶ Ra ²¹⁰ Po	$\begin{array}{c} 0.06(1) \\ 1.3(2) \\ 0.9(2) \end{array}$
Total U/Th α activity		8(2)
	${}^{40}K$ ${}^{60}Co$ ${}^{90}Sr - {}^{90}Y$ ${}^{137}Cs$ ${}^{147}Sm$ ${}^{152}Eu$ ${}^{152}Eu$ ${}^{154}Eu$	$\leq 7 \\ \leq 3 \\ \leq 4 \\ \leq 0.3 \\ 0.34(4) \\ 10(2) \\ \leq 0.5$



2. Fit of the <u>a spectrum</u> (efficiency=68% from ¹⁴⁷Sm peak) in [225,365 keV]: $5 = (374 \pm 329) \text{ counts} \longrightarrow T_{1/2} = 4.4^{+11.3}_{-2.1} (stat) \times 10^{18} \text{ y}$ $E_{peak} = (253\pm7) \text{ keV} \rightarrow (1.96\pm0.03) \text{ MeV}$

¹⁵¹Eu α decay: $T_{1/2} = 5^{+11}_{-3} \times 10^{18}$ y ... or in a conservative approach: $T_{1/2} \ge 1.7 \times 10^{18}$ y (68% C.L.)

α decay of ¹⁵¹Eu to the first excited level of ¹⁴⁷Pm



Subsequent 91 keV deexcitation energy (γ , 33%, or X rays/Auger electrons, 67%) effectively absorbed

 \Rightarrow Expected peak at (325±33) keV



Counts / 5 keV



Fit model:

2 Gaussian peaks (a decay of ¹⁵¹Eu and ¹⁴⁷Sm) + exponential function (background)

No evidence:

$$T_{1/2}(g.s. \rightarrow 5/2^+) \ge 6 \times 10^{17} \,\mathrm{yr}$$

(68% C.L.)

Implications

Presence of Promethium in the Earth crust

- It is one of the rarest elements (all isotopes unstable with $T_{1/2}$ <18 yr)

Present only as temporal product (predominantly from ²³⁸U spontaneous fission)
 It is possible to calculate about 560 g of natural Pm in the Earth crust

Observed a decay of ¹⁵¹Eu opens additional source of natural Pm.

Taking into account the abundance of Europium in the crust (2×10⁻⁴%)

 \Rightarrow additional 12^{+17}_{-8} g of natural Pm born by ¹⁵¹Eu

Comparison with theory

B. Buck et al., J. Phys. G 17 (1991) 1223; 18 (1992) 143.
 D.N. Poenaru, M. Ivascu, J. Physique 44 (1983) 791.
 G. Royer, J. Phys. G 26 (2000) 1149.
 M. Fujiwara et al., J. Phys. G 28 (2002) 643.
 D.N. Poenaru et al., Phys. Rev. C 32 (1985) 2198.

Populated	Theoretical $T_{1/2}$, yr						
level	[1]	[2]	[3]	[4]	[5]	Experimental $T_{1/2}$, yr	
ground state $5/2^+$, 91 keV	3.0×10^{17} 7.7×10^{18}	3.6×10^{18} 1.0×10^{20}	$\begin{array}{c} 6.3 imes 10^{17} \ 1.5 imes 10^{19} \end{array}$	5.9×10^{17} 1.7×10^{19}	$1.6 imes 10^{18}$ _	$5^{+11}_{-3} \times 10^{18}$ $\geq 6 \times 10^{17}$	

G.S.: Measured $T_{1/2}$ is in agreement with theoretical value [2] and [5].

Search for $2\beta 2\nu (0^+ \rightarrow 0^+_1)$ of ¹⁰⁰Mo

in the volume Nucl.Phys. and Atomic Energy, ed. INR-Kiev (2007), 479



Transition (g.s. \rightarrow g.s.) of ¹⁰⁰Mo has been observed in six experiments: $T_{1/2} = 7.7 \pm 0.5 \times 10^{18}$ yr [JETP Letters 80(2004)377]



Transition of ¹⁰⁰Mo to O⁺₁ level of ¹⁰⁰Ru has been positively identified in four experiments by Barabash et al.:

 $T_{1/2} = 6.1^{+1.8} - 1.1 \times 10^{20} \text{ y}$ $T_{1/2} = 9.3^{+2.8} - 1.7 \times 10^{20} \text{ y}$ $T_{1/2} = 5.9^{+1.8} - 1.3 \times 10^{20} \text{ y}$ $T_{1/2} = 5.7^{+2.1} - 1.7 \times 10^{20} \text{ y}$

[Phys.Lett.B 345(1995)408] [Phys.At.Nucl. 62(1999)2039] [Phys.Rev.Lett. 86(2001)3510] [Nucl.Phys.A 781(2007)209]

in agreement among themselves, but not with the old NEMO data: $T_{1/2} > 1.2 \times 10^{21}$ y at 90% C.L. [Phys.Lett.B 275(1992)506]

Experimental set-up



in the volume Nucl.Phys. and Atomic Energy, ed. INR-Kiev (2007), 479

ARMONIA Collaboration:

meAsuReMent of twO-NeutrIno ββ decAy of ¹⁰⁰Mo to the first excited O* level of ¹⁰⁰Ru

Aim of the experiment: to confirm previous positive results or to set more stringent limit on the probability of the process

Data collected at LNGS in low-background set-up with 4 HP Ge detectors (each of ~225 cm³).

Mo sample:

1009 g of Mo (metallic powder) enriched by ¹⁰⁰Mo at 99.5%, compressed to ~6 g/cm³ to fill fully the well in the HP Ge set-up; some amount also above the detectors.



Germanium detectors parameters				
	Detecto	rs		
	ge178	ge179	ge180	ge188
Volume (cm ³)	225.2	225.0	225.0	220.7
Endcap and holder material	Electrol	ytical cop	per	
Energy resolution (FWHM) at 1332 keV	2.1	2.0	2.0	2.0



DATA TAKING IN PROGRESS

Search for $\beta\beta$ processes in ⁶⁴Zn

Preprint: ROM2F/2007/13

Low background ZnWO₄ scintillator, 20 × 19 × 40 mm -> 117 g mass
 Inside a cavity (filled up with high-pure silicon oil) \$\phi 47 × 59 mm in central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length.
 2 Low background photomultiplier (PMT) EMI9265-B53/FL 3" diameter.

(All light guide wrapped by PTFE reflection tape)



Search for $\beta\beta$ processes in ⁶⁴Zn

Preprint: ROM2F/2007/13

Big natural abundance: 48.268%, M(⁶⁴Zn) - M(⁶⁴Ni) = 1095.7(0.7)keV



[10] H.J. Kim et al., KEK Proc. 6 (2003) 205; report on the Int. Conf. on Current

Problems in Nucl. Phys. At. Energy, Kiyv, Ukraine, May 29 { June 3, 2006.

[18] T. Bloxham et al., arXiv: 0707.2756v1 [nucl-ex].

Conclusions

Many rare processes have been investigated by DAMA coll. by developing and using various low background scintillators and set-ups increasing the sensitivities to various processes.

Here just results published in 2007 have been summarised.

- 1. New limits on α decay modes with a Li₆Eu(BO₃)₃ crystal
- 2. First measurement of α decay of ¹⁵¹Eu with CaF₂(Eu) detector
- 3. Preliminary results and improvement of the experiment to study $2\beta 2\nu(0^+ \rightarrow 0^+_1)$ of ¹⁰⁰Mo with ~1 kg Mo sample.
- 4. New limits on 2 β processes in ⁶⁴Zn with ZnWO₄

Further works in progress and in preparation

Properties – LaCl ₃ (Ce)
Density [g/cm³] 3.79
Melting point [K] 1135
Thermal expansion coefficient [10 ⁻⁶ /°C] 11 along C-axis
Cleavage plane [100]
Hygroscopicyes
Wavelength of emission max. [nm] 350
Lower wavelength cutoff [nm]
Refractive index @ emission max ~1.9
Primary decay time [µs]0.028
Light yield [photons/keVγ]49
Photoelectron yield [% of NaI(Tl)] (for γ- rays)

Properties – CaF ₂ (Eu)
Density [g/cm³] 3.18
Melting point [K] 1691
Thermal expansion coefficient [C ⁻¹]19.5x10 ⁻⁶
Cleavage plane
Hardness (Mho) 4
Hygroscopic no
Wavelength of emission max. [nm] 435
Lower wavelength cutoff [nm]
Refractive index @ emission max 1.47
Primary decay time [μs] 0.94
Light yield [photons/keVγ]19
Photoelectron yield [% of NaI(Tl)] (for γ-rays) 50

Mass-spectrometry of the $CaF_2(Eu)$ crystal

Concentration of Eu determined with the ICP-MS (Inductively Coupled Plasma - Mass Spectrometry analysis).

Representative part of the $CaF_2(Eu)$ crystal reduced to powder by mechanical treatment inside a cleaned polyethylene bag to avoid possible contamination.

Calcium fluoride rather insoluble in acids: several tests applying different solutions. The best solutions were as following: (a) H2SO4 95%; (b) mixture of 2 parts of HCl and 5 parts of HNO₃. Solubility of CaF_2 was about 75% and 65%, respectively.

The estimated effect of insoluble EuF_3 formation is negligible.

Six solutions were measured by ICP-MS analysis (3 of b, and 3 of a type): average value = (0.4 ± 0.1) % (uncertainty takes into account the uncertainty on the sample preparation procedure).

The contamination by Nd, Sm, Gd, Hf, Os, Pt, Bi, Th, U (all these elements have α active isotopes) were also measured: <0.1 ppm were obtained for all of them.

Energy dependence of the α/β ratio

Because quenching of the scintillation light caused by α particles (in comparison with electrons) is due to the higher ionization density of α particles, such a behaviour of the α/β ratio can be explained by the energy dependence of the ionization density of α particles.

Increasing of α/β ratio at energies of α particles lower than 1.5 – 2 MeV was observed for CdWO₄ [1], and CaWO4 [2] scintillators. Similar behaviour of α/β ratio was measured very precisely for CsI(Tl) scintillator [3].

[1] F.A. Danevich et al., Phys. Rev. C 67 (2003) 014310.
[2] Yu.G. Zdesenko et al., Nucl. Instrum. Meth. A 538 (2005) 657.
[3] R. Gwin, R.B. Murray, Phys. Rev. 131 (1963) 501.

Fit was performed in the energy region with starting point from **215 to 230 keV** (with step of 5 keV) and with final point from **340 to 410 keV**. In the most of the intervals the fit gives positive effect at the level of **1.2 - 2.8** σ .

The best fit (χ^2 /n.d.f. = 0.42), achieved in the interval 225 – 365 keV: S = (302±232) counts.

Feldman-Cousins procedure \Rightarrow it is possible to conclude that observation of the effect is statistically significant and amplitude of the effect is inside the interval [5, 682] counts with 90% C.L. (or [98, 534] @ 68% C.L.).

 $T_{1/2} = 5.4^{+11.4}_{-2.4} (stat) \pm 1.4 (syst) \times 10^{18} \text{ y}$

The systematic error is related mainly with the uncertainty of the number of Eu nuclei in the crystal and with variation of the peak's area depending on the interval of the fit.

There are several α decaying nuclides with energy releases close to the ^{151}Eu value:

- ¹⁴⁴Nd (Q_{α} = 1.905 MeV), • ¹⁴⁸Sm (Q_{α} = 1.986 MeV) and
- ${}^{152}Gd(Q_{a}=2.203 \text{ MeV})$

¹⁴⁸Sm has near the same abundance as ¹⁴⁷Sm; however, because of its big halflife (7×10¹⁵ y in comparison with 1.06×10¹¹ y for ¹⁴⁷Sm), <u>peak of ¹⁴⁸Sm should be</u> <u>near five orders of magnitude lower than peak of ¹⁴⁷Sm</u> and, thus, cannot mimic alpha decay of ¹⁵¹Eu.

Contaminations of the CaF₂(Eu) crystal by Nd and Gd are lower than 0.1 ppm that is known from the ICP-MS analysis. With $T_{1/2} = 2.29 \times 10^{15}$ y and $\delta = 23.8\%$, ¹⁴⁴Nd could contribute not more than 10 events in the ¹⁵¹Eu peak.

<u>Contribution of ¹⁵²Gd</u> ($T_{1/2}$ = 1.08 × 10¹⁴ y and δ = 0.2%) <u>should be even lower</u>.

Thus, the observed alpha peak at \approx 250 keV cannot be mimicked by some other α decaying nuclides present in the CaF₂(Eu) crystal.

