

**PRELIMINARY RESULTS ON THE SEARCH FOR  $^{100}\text{Mo}$   $2\beta$  DECAY  
TO THE FIRST EXCITED  $0^+_1$  LEVEL OF  $^{100}\text{Ru}$**

**P. Belli<sup>1</sup>, R. Bernabei<sup>1</sup>, R. S. Boiko<sup>2</sup>, R. Cerulli<sup>3</sup>, F. A. Danevich<sup>4</sup>, S. d'Angelo<sup>1</sup>, A. Incicchitti<sup>5</sup>,  
V. V. Kobychyev<sup>4</sup>, B. N. Kropivnyansky<sup>4</sup>, M. Laubenstein<sup>4</sup>, P. G. Nagorny<sup>b</sup>, S. S. Nagorny<sup>4</sup>,  
S. Nisi<sup>3</sup>, D. V. Poda<sup>4</sup>, D. Prospero<sup>5</sup>, V. I. Tretyak<sup>4</sup>, I. M. Vyshnevskiy<sup>4</sup>, S. S. Yurchenko<sup>4</sup>  
(ARMONIA\* Collaboration)**

<sup>1</sup> *Dipartimento di Fisica, Universita' di Roma "Tor Vergata" and INFN, Rome, Italy*

<sup>2</sup> *Chemical department, Kyiv National Taras Shevchenko University, Kyiv, Ukraine*

<sup>3</sup> *INFN - Laboratori Nazionali del Gran Sasso, Italy*

<sup>4</sup> *Institute for Nuclear Research, National Academy of Sciences of Ukraine, Kyiv, Ukraine*

<sup>5</sup> *Dipartimento di Fisica, Universita' di Roma "La Sapienza" and INFN, Rome, Italy*

Molybdenum sample with mass of near 1 kg, enriched in  $^{100}\text{Mo}$  at 99,5 %, is used to confirm or reject previous reports on observation of  $2\beta 2\nu$  decay of  $^{100}\text{Mo}$  to the first excited  $0^+_1$  level of  $^{100}\text{Ru}$  at  $E(0^+_1) = 1130,5$  keV. Data are collected in the low-background set-up with 4 HP Ge detectors, each of 225 cm<sup>3</sup> volume, in the Gran Sasso Underground Laboratory (Italy). After 1927 h of measurements, the reached sensitivity to the half-life is  $\sim 5 \cdot 10^{20}$  y; it does not allow to make definite conclusions on existence of the process searched for. Further measurements are in progress.

### 1. Introduction

The great interest to the double beta ( $2\beta$ ) decay search [1 - 8] is related, in particular, with the recent evidence for neutrino oscillations, which strongly suggests that neutrinos have nonzero mass. While oscillation experiments are sensitive only to the neutrino mass difference, measurement of neutrinoless ( $0\nu$ ) double beta decay could define the nature of the neutrinos (Majorana or Dirac) and the absolute scale of the effective neutrino mass. Neutrinoless mode of  $2\beta$  decay is forbidden in the Standard Model (SM) as violating the lepton number conservation. At the same time, two-neutrino ( $2\nu$ ) mode is fully allowed in the SM. However, as a second-order weak process,  $2\beta 2\nu$  decay occurs at extremely low rates. To date, this process was observed in direct experiments only for 7 nuclei ( $^{48}\text{Ca}$ ,  $^{76}\text{Ge}$ ,  $^{82}\text{Se}$ ,  $^{96}\text{Zr}$ ,  $^{100}\text{Mo}$ ,  $^{116}\text{Cd}$ , and  $^{150}\text{Nd}$ ) with half-lives in the range of  $\sim 10^{19}$ – $10^{21}$  y (see f.e. [4]) that makes it currently the rarest process of radioactive decay. While being an interesting by itself, the  $2\beta 2\nu$  decay is considered also as a tool for checking the calculations of nuclear matrix elements inside different theoretical approaches [5, 9, 10] that is important for

extraction of reliable values of the effective neutrino mass from the measurements of the neutrinoless mode of  $2\beta$  decay.

For two nuclides,  $^{100}\text{Mo}$  and  $^{150}\text{Nd}$ ,  $2\beta 2\nu$  decay was observed not only for transitions to the ground states, but also to the first excited  $0^+_1$  levels of daughter nuclei. Here we discuss the  $^{100}\text{Mo}$  observations and possibilities to check and improve the accuracy of the reported results with a sample of molybdenum enriched by  $^{100}\text{Mo}$  at 99,5 %<sup>†</sup>, which belongs to the Kiev Institute for Nuclear Research, in underground measurements in the Laboratorio Nazionale del Gran Sasso with low-background HP Ge detectors available at LNGS.

Scheme of  $^{100}\text{Mo}$   $2\beta$  decays is shown in Fig. 1. Full energy release for the ground state (g.s.) to g.s. transition is equal  $3035 \pm 6$  keV [13]. Two-neutrino double beta decay of  $^{100}\text{Mo}$  to the  $^{100}\text{Ru}$  ground state was observed in 6 experiments with the most recent

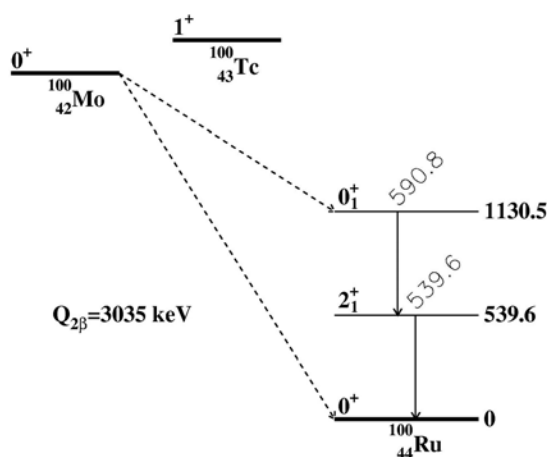


Fig. 1. Scheme of  $2\beta$  decay of  $^{100}\text{Mo}$  to the ground state and to the first  $0^+_1$  excited level of  $^{100}\text{Ru}$ . Energies of levels and de-excitation  $\gamma$  quanta are given in keV [12].

\* MeAsuReMent of twO-NeutrIno  $\beta\beta$  decAy of  $^{100}\text{Mo}$  to the first excited  $0^+$  level of  $^{100}\text{Ru}$ .

<sup>†</sup> Natural abundance of  $^{100}\text{Mo}$  is 9.63% [11].

and accurate value of  $T_{1/2}^{2\nu}(\text{g.s.} \rightarrow \text{g.s.}) = (7,7 \pm 0,5) \cdot 10^{18}$  y given by the NEMO-3 measurements [14] (other  $T_{1/2}$  results are in agreement with this value, see data and references in [4]). In two-neutrino  $2\beta$  decay of  $^{100}\text{Mo}$  to the first excited  $0^+_1$  level of  $^{100}\text{Ru}$ , in addition to two electrons, two deexcitation  $\gamma$  quanta with energies of  $E(\gamma_1) = 590,8$  keV and  $E(\gamma_2) = 539,6$  keV are also emitted (see Fig. 1). This process was positively identified in four experiments:

1) measurements with  $\sim 1$  kg sample of  $^{100}\text{Mo}$  performed in the Soudan mine (2090 m w.e.) with  $114 \text{ cm}^3$  HP Ge detector during 415 d:  $T_{1/2}^{2\nu}(\text{g.s.} \rightarrow 0^+_1) = 6,1^{+1,8}_{-1,1} \cdot 10^{20}$  y [15];

2) studies with a set of  $^{100}\text{Mo}$  enriched metal powder samples and few low-background HP Ge detectors in the Modane Underground Laboratory (4800 m w.e.); data from 17 measurements gave:  $T_{1/2}^{2\nu}(\text{g.s.} \rightarrow 0^+_1) = 9,3^{+2,8}_{-1,7} \cdot 10^{20}$  y [16];

3) 440 d measurements with two HP Ge detectors ( $280 \text{ cm}^3$  each) in coincidence in the earth-based Low Background Counting Facility of the Triangle Universities Nuclear Laboratory:  $T_{1/2}^{2\nu}(\text{g.s.} \rightarrow 0^+_1) = 5,9^{+1,8}_{-1,3} \times 10^{20}$  y [17];

4) preliminary result of the NEMO-3 measurements (which are still in progress) with 7 kg of  $^{100}\text{Mo}$  during  $\sim 1$  y in the NEMO-3 tracking detector [18]:  $T_{1/2}^{2\nu}(\text{g.s.} \rightarrow 0^+_1) \cong 6 \cdot 10^{20}$  y [19].

However, while being in agreement between themselves, these results are in contradiction (at  $3,3 \sigma$  for more accurate results [15, 17]) with data of the fifth experiment which was performed also in the Modane Underground Laboratory with  $\sim 1$  kg molybdenum sample, enriched in  $^{100}\text{Mo}$  at 99,5 %, and  $100 \text{ cm}^3$  HP Ge detector during 96 d, where only the limit was found:  $T_{1/2}^{2\nu}(\text{g.s.} \rightarrow 0^+_1) > 1,2 \cdot 10^{21}$  y at 90 % C.L. [20].

Aim of the present work is to remeasure this sample of  $^{100}\text{Mo}$  during bigger time and with HP Ge detectors of bigger volume in the low-background conditions of the Laboratori Nazionali del Gran Sasso (LNGS) to confirm previous positive results of Refs. [15 - 17, 19] or to set more stringent limit on the probability of this process than that established in Ref. [20].

## 2. Measurements and results

Data are collected deep underground (3600 m w.e.) at LNGS in the low-background set-up with 4 HP Ge detectors (each of  $\sim 225 \text{ cm}^3$ ). Detectors are mounted altogether in one cryostat; energy resolution of the detectors is 2 keV at 1332 keV line of  $^{60}\text{Co}$ . This apparatus was recently successfully used in an experiment with 929 g sample of In, where in measurements during 115 d very rare single  $\beta$  decay of  $^{115}\text{In}$  (which constitutes 95,7 % of natural In) to the first excited level of  $^{115}\text{Sn}$  ( $E_{\text{exc}} = 497$  keV) was observed at the first time with  $4\sigma$  confidence level [21]. The measured half-life was equal  $T_{1/2} = 3,7 \cdot 10^{20}$  y. Thus, the sample mass, the  $\gamma$  quanta energy and sensitivity of this set-up were just those needed for measurements with the  $^{100}\text{Mo}$  sample.

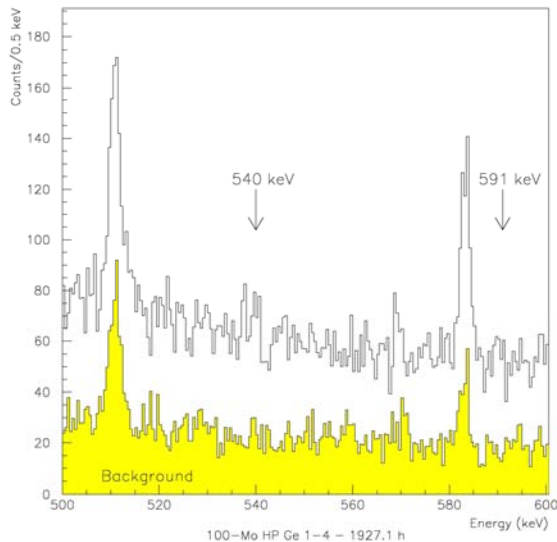


Fig. 2. Spectrum of  $^{100}\text{Mo}$  sample (mass of 1009 g) measured with 4 HP Ge detectors set-up at LNGS during 1927 h in the range of 500 - 600 keV. Shaded area is background spectrum (without  $^{100}\text{Mo}$  sample) normalised to the same time of measurements. Peaks at 511 keV and 583 keV are related with  $^{208}\text{Tl}$  decay and annihilation process (511 keV).

Measurements [20] showed that pollution of the  $^{100}\text{Mo}$  powder by radioactive nuclides was on the level of (in mBq/kg):  $^{232}\text{Th}$  family  $\sim 2$ ;  $^{235}\text{U}$  family  $\sim 40$ ;  $^{238}\text{U}$  family  $\sim 10$  ( $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ ) and  $\sim 500$  ( $^{234}\text{Th}$ ,  $^{234\text{m}}\text{Pa}$ );  $^{40}\text{K} \sim 200$ ;  $^{137}\text{Cs} \sim 8$ ; and  $^{207}\text{Bi} \sim 0,5$ . It was concluded that, to reach high sensitivity, the Mo sample should be purified. Purification was performed at the Chemical department of the Kyiv National Taras Shevchenko University taking into account experience of Mo purification in the NEMO experiment [22]. However, in this first attempt the aim was not reached, and, as revealed further measurements, in fact the Mo sample was even slightly polluted. In the pilot run of measurements, this Mo specimen was used.

The  $^{100}\text{Mo}$  sample consisted of 1009 g of Mo in form of metallic powder. It was compressed to the density of near  $6 \text{ g/cm}^3$  to fill fully the well in the HP Ge set-up; some amount of Mo (which was outside the capacity of the well) was located above the detectors. Data were collected during 1927 h.

In accordance with the scheme of  $^{100}\text{Mo}$  decay (see Fig. 1), after population of the  $0^+_1$  level of  $^{100}\text{Ru}$ , two gamma quanta with energies of  $E(\gamma_1) = 590,8$  keV and  $E(\gamma_2) = 539,6$  keV are emitted in the de-excitation process. Efficiency to detect these gammas with the HP Ge detectors was calculated with GEANT4 [23] as  $\sim 2,5\%$ . The  $^{100}\text{Mo}$  spectrum in the energy range of 500 - 600 keV is presented in Fig. 2 in comparison with background spectrum without sample. It is evident that counting rate of the Mo sample is  $\sim 2,5$  times higher than that of background in this region due to the Mo pollution.

In the  $^{100}\text{Mo}$  spectrum, one can see a peak structure at energy of 540 keV – just at the place where one of gamma quanta searched for is expected. If to suppose that this peak is related with  $2\beta 2\nu$  decay of  $^{100}\text{Mo}$  to  $0^+_1$  level of  $^{100}\text{Ru}$ , corresponding half-life value is equal  $T_{1/2} = 3 \cdot 10^{20}$  y. However, at the energy of 591 keV, where the second peak should be located, there is no significant statistical evidence of its existence, and only limit on half-life can be given as  $T_{1/2} > 6 \cdot 10^{20}$  y at 90 % C.L.

### 3. Conclusions

In this paper the results of pilot measurements with 1 kg Mo source (enriched by  $^{100}\text{Mo}$  at 99,5 %) at the Gran Sasso National Laboratory of I.N.F.N. with 4 HP Ge detectors have been presented. After 1927 h of data taking, sensitivity of the experiment in terms of  $^{100}\text{Mo}$  half-life for  $2\beta 2\nu$  decay to  $0^+_1$  level of  $^{100}\text{Ru}$  is on the level of  $\sim 5 \cdot 10^{20}$  y. Collected statistics and not sufficient purity of the Mo sample does not currently allow to make definite conclusions about the process. Recently, we performed further chemical purification of the  $^{100}\text{Mo}$  source at LNGS. Data taking with the new sample is in progress. After 1 y of measurements, expected sensitivity will be on the level of  $\sim 3 \cdot 10^{21}$  y that will allow to confirm or reject previous reports on positive observation of  $^{100}\text{Mo}$   $2\beta 2\nu$  decay to  $0^+_1$  level of  $^{100}\text{Ru}$ .

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### ПРЕДВАРИТЕЛЬНЫЕ РЕЗУЛЬТАТЫ ПОИСКА $2\beta$ -РАСПАДА $^{100}\text{Mo}$ НА ПЕРВЫЙ $0^+_1$ ВОЗБУЖДЕННЫЙ УРОВЕНЬ $^{100}\text{Ru}$

**П. Белли, Р. Бернабей, Р. С. Бойко, Р. Черулли, Ф. А. Даневич, С. д'Анджело, А. Инчикитти,  
В. В. Кобычев, Б. Н. Кропивянский, М. Лаубенштейн, П. Г. Нагорный, С. С. Нагорный, С. Ниси,  
Д. В. Пода, Д. Проспери, В. И. Третьяк, И. Н. Вишневский, С. С. Юрченко**

Образец молибдена массой около 1 кг, на 99,5 % обогащенный  $^{100}\text{Mo}$ , используется для подтверждения или опровержения предыдущих сообщений о наблюдении  $2\beta 2\nu$  распада  $^{100}\text{Mo}$  на первый возбужденный  $0^+_1$  уровень

$^{100}\text{Ru}$  с энергией  $E(0^+_1) = 1130,5$  кэВ. Эксперимент проводится в подземной лаборатории Гран Сассо (Италия) в низкофоновой установке с 4 HP Ge детекторами с объемом  $225 \text{ см}^3$  каждый. После измерений длительностью 1927 ч чувствительность к периоду полураспада равна  $\sim 5 \cdot 10^{20}$  лет, что не позволяет сделать окончательные заключения о существовании искомого процесса. Измерения продолжаются.

**ПОПЕРЕДНІ РЕЗУЛЬТАТИ ПОШУКУ  $2\beta$ -РОЗПАДУ  $^{100}\text{Mo}$   
НА ПЕРШІЙ  $0^+_1$  ЗБУДЖЕНИЙ РІВЕНЬ  $^{100}\text{Ru}$**

**П. Беллі, Р. Бернабей, Р. С. Бойко, Р. Черуллі, Ф. А. Даневич, С. д'Анджело, А. Інчікитті,  
В. В. Кобичев, Б. М. Кропив'янський, М. Лаубенштейн, П. Г. Нагорний, С. С. Нагорний, С. Нісі,  
Д. В. Пода, Д. Проспері, В. І. Третьак, І. М. Вишневський, С. С. Юрченко**

Зразок молібдену масою біля 1 кг, на 99,5 % збагаченого  $^{100}\text{Mo}$ , використовується для підтвердження або спростування попередніх повідомлень про спостереження  $2\beta 2\nu$  розпаду  $^{100}\text{Mo}$  на перший збуджений  $0^+_1$  рівень  $^{100}\text{Ru}$  з енергією  $E(0^+_1) = 1130,5$  кеВ. Експеримент проводиться в підземній лабораторії Гран Сассо (Італія) в низькофоновій установці з 4 HP Ge детекторами об'ємом  $225 \text{ см}^3$  кожний. Після вимірювань протягом 1927 год чутливість до періоду напіврозпаду дорівнює  $\sim 5 \cdot 10^{20}$  років, що не дозволяє зробити остаточні висновки про існування шуканого процесу. Вимірювання продовжуються.