universe

MDPI

Communication

Recent Developments and Results on Double Beta Decays with Crystal Scintillators and HPGe Spectrometry †

Alessandro Di Marco [1,*], Alexander S. Barabash [2], Pierluigi Belli [1,3], Rita Bernabei [1,3], Roman S. Boiko [4,5], Viktor B. Brudanin [6], Fabio Cappella [7,8], Vincenzo Caracciolo [9], Riccardo Cerulli [1,3], Dmitry M. Chernyak [4,10], Fedor A. Danevich [4], Antonella Incicchitti [7,8], Dmytro V. Kasperovych [4], Vladislav V. Kobychev [4], Sergey I. Konovalov [2], Matthias Laubenstein [9], Vittorio Merlo [1,3], Francesco Montecchia [1,11], Oksana G. Polischuk [4], Denys V. Poda [4,12], Vladimir N. Shlegel [13], Vladimir I. Tretyak [4], Vladimir I. Umatov [2], Yan V. Vasiliev [13], and Mykola M. Zarytskyy [4]

1 Istituto Nazionale di Fisica Nucleare, Sezione di Roma "Tor Vergata", 00133 Rome, Italy; pierluigi.belli@roma2.infn.it (P.B.); rita.bernabei@roma2.infn.it (R.B.); riccardo.cerulli@roma2.infn.it (R.C.); vittorio.merlo@roma2.infn.it (V.M.); francesco.montecchia@uniroma2.it (F.M.)
2 National Research Centre "Kurchatov Institute", Institute of Theoretical and Experimental Physics, 123182 Moscow, Russia; barabash@itep.ru (A.S.B.); konovalov@itep.ru (S.I.K.); umatov@itep.ru (V.I.U.)
3 Dipartimento di Fisica, Università di Roma "Tor Vergata", 00133 Rome, Italy
4 Institute for Nuclear Research, National Academy of Sciences of Ukraine, 03680 Kyiv, Ukraine; boiko@kinr.kiev.ua (R.S.B.); chernyak@kinr.kiev.ua (D.M.C.); danevich@kinr.kiev.ua (F.A.D.); casper.phys@gmail.com (D.V.K.); kobychev@kinr.kiev.ua (V.V.K.); polischuk@kinr.kiev.ua (O.G.P.); denys.poda@csnsm.in2p3.fr (D.V.P.); vladimir.tretyak.vit@gmail.com (V.I.T.); zaritsky96@gmail.com (M.M.Z.)
5 Department of Organic, Physical and Colloid Chemistry and Chemistry of Pesticides, National University of Life and Environmental Sciences of Ukraine, 03041 Kyiv, Ukraine
6 Institute for Nuclear Research, 141980 Dubna, Russia; brudanin@jinr.ru
7 Dipartimento di Fisica, Università di Roma "La Sapienza", 00185 Rome, Italy; fabio.cappella@roma1.infn.it (F.C.); antonella.incicchitti@roma1.infn.it (A.I.)
8 Istituto Nazionale di Fisica Nucleare, Sezione di Roma, 00185 Rome, Italy
9 Laboratori Nazionali del Gran Sasso, Istituto Nazionale di Fisica Nucleare, 67100 Assergi, Italy; vincenzo.caracciolo@lngs.infn.it (V.C.); matthias.laubenstein@lngs.infn.it (M.L.)
10 Kavli Institute for the Physics and Mathematics of the Universe (WPI), University of Tokyo, Kashiwa 277-8583, Japan
11 Dipartimento di Ingegneria Civile e Ingegneria Informatica, Università di Roma "Tor Vergata", 00133 Rome, Italy
12 Centre de Sciences Nucléaires et de Sciences de la Matiére, Université Paris-Sud, CNRS/IN2P3, Université Paris-Saclay, 91405 Orsay, France
13 Nikolaev Institute of Inorganic Chemistry, 630090 Novosibirsk, Russia; shlegel@niic.nsc.ru (V.N.S.); yan.vasiliev@gmail.com (Y.V.V.)
* Correspondence: alessandro.dimarco@roma2.infn.it
† This paper is based on the talk at the 7th International Conference on New Frontiers in Physics (ICNFP 2018), Crete, Greece, 4–12 July 2018.

Received: 3 November 2018; Accepted: 6 December 2018; Published: 14 December 2018

Abstract: Recent developments, results, and perspectives arising from double beta decay experiments at the Gran Sasso National Laboratory (LNGS) of the INFN by using HPGe detectors and crystal scintillators and by exploiting various approaches and different isotopes are summarized. The measurements here presented have been performed in the experimental set-ups of the DAMA collaboration. These setups are optimized for low-background studies and operate deep underground at LNGS. The presented results are of significant value to the field, and the sensitivity achieved for some of the considered isotopes is one of the best available to date.

Keywords: double beta decay; scintillators; low background measurements; rare processes

1. Introduction

DAMA is a pioneer project for the investigation of dark matter (DM), and it is also very active in the development of new highly radiopure crystal scintillators for their application to the search for rare processes. Many significant results have been obtained by investigating various rare processes in many experiments performed at the Gran Sasso National Laboratory (LNGS) by DAMA and its collaboration with researchers from INR-Kyiv and other institutions.

The most recent results obtained from the Large sodium Iodide Bulk for Rare processes DAMA/LIBRA-phase2 investigation of DM combined with those from the DAMA/NaI and DAMA/LIBRA-phase1 setups are presented elsewhere [1].

Here, some of the main results obtained from the search for rare processes with the DAMA setups[1] are briefly discussed; some details on the observation of $2\nu2\beta$ decays from the meAsuReMent of twO-NeutrIno $\beta\beta$ decAy of ^{100}Mo to the first excited 0^+ level of ^{100}Ru (ARMONIA) [2] and AURORA [3] experiments are given, and the latest activities on ^{106}Cd and ^{150}Nd [4] double beta decay are presented.

Many results have been obtained with the DAMA setups in experiments investigating the double beta decay of several candidate isotopes at LNGS; in particular, double beta decay processes in the following isotopes were investigated: ^{40}Ca, ^{46}Ca, ^{48}Ca, ^{64}Zn, ^{70}Zn, ^{100}Mo, ^{96}Ru, ^{104}Ru, ^{106}Cd, ^{108}Cd, ^{114}Cd, ^{116}Cd, ^{112}Sn, ^{124}Sn, ^{134}Xe, ^{136}Xe, ^{130}Ba, ^{136}Ce, ^{138}Ce, ^{142}Ce, ^{150}Nd, ^{156}Dy, ^{158}Dy, ^{162}Er, ^{170}Er, ^{180}W, ^{186}W, ^{184}Os, ^{192}Os, ^{190}Pt, and ^{198}Pt. The sensitivities achieved for the half-life of the studied processes are competitive (between 10^{20} and 10^{24} year) due to the radiopurity of the detectors developed and the experimental approaches used. The results have improved (often by several orders of magnitude) the half-life limits obtained by previous experiments and have enabled new observations of two-neutrino double beta decay of ^{100}Mo [2], ^{116}Cd [3], and, preliminarily, ^{150}Nd [4]. Moreover, the obtained experimental sensitivities to decay modes with positron emission or double electron capture for some of the candidate isotopes are the best in the field.

As regards the rare α and β decays, we have obtained the first observation of ^{151}Eu α decay with $T_{1/2} = 5 \times 10^{18}$ year through the use of a CaF_2 (Eu) crystal scintillator [5]; we have also achieved the α decay of ^{190}Pt to the first excited level ($E_{exc} = 137.2$ keV) of ^{186}Os with $T_{1/2} = 3 \times 10^{14}$ year [6]. The rare β decays of ^{113}Cd and ^{48}Ca have been investigated using $CdWO_4$ [7] and CaF_2 (Eu) [8] crystal scintillators, respectively. Moreover, pairs of NaI (Tl) detectors of the DAMA/LIBRA setup have been used to search for production of correlated e^+e^- pairs in the α decay of ^{241}Am [9].

Solar axions have been sought by studying their conversion to photons (inverse Primakoff effect) in NaI (Tl) crystals [10] and by investigating the resonance excitation of the ^{7}Li nuclei in a LiF crystal [11] and Li-containing powders [12]; the latter approach was based on the hypothetical axions emitted in the de-excitation of ^{7}Li nuclei in the Sun. Delayed coincidences have been investigated to search for such exotic particles as Q-balls [13] and SIMPs [14] using the DAMA/NaI detectors, and DAEMONs have been studied using the specially developed NEMESIS setup [15]. Electron stability has been investigated by searching for electron "disappearance" (i.e., decay into invisible

---

1 DAMA operates several low-background setups at LNGS: DAMA/NaI (out of operation in 2002), DAMA/LIBRA, DAMA/R&D, DAMA/CRYS, DAMA/LXe (out of operation in 2018), DAMA/Ge, and other HPGe detectors from the STELLA facility.

channels as $e^- \rightarrow \nu_e \bar{\nu}_e \nu_e$) [16,17] and by searching for the $e^- \rightarrow \nu_e \gamma$ decay mode [17,18]. Finally, competitive limits have been obtained on the lifetime of several other possible nuclear processes. In particular, the following have been studied: (i) the spontaneous transition of ^{23}Na and ^{127}I nuclei to a superdense state [19]; (ii) cluster decays of ^{127}I [20] and of ^{138}La and ^{139}La [21]; (iii) nucleon, di-nucleon, and tri-nucleon decay into invisible channels [22,23]; (iv) Charge non-conserving (CNC) processes in ^{127}I [24]; (v) CNC β decay of ^{136}Xe [23], ^{100}Mo [2], and ^{139}La [25]; (vi) CNC electron capture with nuclear-level excitation in ^{127}I and ^{23}Na [26] and in ^{129}Xe [27]; (vii) nuclear processes violating the Pauli exclusion principle in sodium and iodine [28,29]; (viii) several rare nuclear decays in a BaF_2 crystal scintillator contaminated by radium [30]; (ix) long-lived superheavy ekatungsten with a radiopure $ZnWO_4$ crystal scintillator [31].

2. Observation of $2\nu2\beta$ Decay of ^{100}Mo in the ARMONIA Experiment

To date, among the 35 naturally occurring $2\beta^-$ candidates [32], more than 10 have been experimentally observed undergoing this process. One of the most interesting isotopes that has been the subject of 2β decay investigation is ^{100}Mo. The interest in this isotope is due to several aspects, including the following: (i) its natural abundance is rather high: $\delta = 9.744(65)\%$ [33]; (ii) it has a high energy release, $Q_{2\beta} = 3034.36(17)$ keV [34], which yields a large phase space integral of the decay and thus a relatively high probability of the occurrence of 2β decay processes; moreover, this $Q_{2\beta}$ value is even higher than the 2615 keV γ line from ^{208}Tl, which represents the highest-energy γ line from natural radioactivity (mostly ^{238}U, ^{232}Th, and ^{40}K), leading to lower achievable background; (iii) there is a possibility to obtain isotopically enriched material by using comparatively inexpensive ultra-speed centrifuge technology.

The half-life of the ^{100}Mo 2β decay isotope has been measured by a geochemical experiment [35] and by several direct experiments in which the $2\nu2\beta$ decay to the ground state of ^{100}Ru was observed with $T_{1/2}$ values in the range of (3.3–11.5) $\times$ 10^{18} year [32,36,37].

The $2\nu2\beta$ decay of ^{100}Mo was also registered for the transition to the first excited 0^+_1 level of ^{100}Ru, and the half-lives were measured in several experiments [38–46] in the range: (5.5–9.3) $\times$ 10^{20} year. However, these positive evidences are in conflict with an earlier result [47], which gave only the limit $T_{1/2} > 1.2 \times 10^{21}$ year at 90% C.L.

The aim of the ARMONIA experiment at LNGS's underground laboratories [2] was to remeasure $\simeq$1 kg of Mo enriched in ^{100}Mo to 99.5%, used in [47], with more measurements and higher sensitivity in order to confirm the observations reported in [38–46] or to set an even more stringent $T_{1/2}$ limit. If the 0^+_1 excited level of ^{100}Ru ($E_{exc} = 1130.3$ keV) is populated, two γs with energies of 590.8 keV and 539.5 keV will be emitted in cascade in the resulting de-excitation process. These γs have been searched for using the GeMulti setup. This setup is equipped with four low-background HPGe detectors mounted in one cryostat with a well in the center; the HPGe detectors have volumes of 225.2, 225.0, 225.0, and 220.7 cm^3, respectively. The typical energy resolution (FWHM) of the detectors is 2.0 keV at the 1332 keV line of ^{60}Co. A lead and copper passive shield surrounds the experimental setup and has a nitrogen ventilation system to avoid radon near the detectors. A sample of metallic ^{100}Mo powder with a mass of 1009 g and a 99.5% enrichment in ^{100}Mo was measured at the first stage of the experiment. The collected data indicated the occurrence of the sought-after 2β decay [48]. Then, to reduce the background counting rate for the sample, it was further purified of radioactive residual contaminants. The ^{100}Mo metal was transformed into molybdenum oxide ($^{100}MoO_3$) with a mass of 1199 g. The purification procedure effectively removed ^{40}K and ^{137}Cs, and it also led to a reduction in the U/Th concentration [49]. The obtained sample of $^{100}MoO_3$ was measured for 18120 h in the GeMulti setup. The background of the setup was collected under the same running conditions as the sample before (for 3211 h) and after (for 4500 h) the measurements with the sample to obtain consistent results; thus, in total, the background was measured over 7711 h.

The one-dimensional energy spectra measured with the $^{100}MoO_3$ sample and the background in the (490–630) keV energy region are shown in Figure 1, left. Two peaks 540 keV and 591 keV (expected for ^{100}Mo $\rightarrow$ ^{100}Ru(0^+_1) $2\nu2\beta$ decay) were observed in the experimental data collected with the ^{100}MoO$_3$ sample, while these peaks are absent in the background spectrum. The ^{100}MoO$_3$ had mass of 1199 g and 99.5% enrichment in ^{100}Mo; thus, it contained $N = 4.85 \times 10^{24}$ ^{100}Mo nuclei. The number of events in the 539.5 keV peak was determined by fitting the experimental energy spectrum to the energy interval (480–560) keV by the sum of the exponential distribution (which represents the background) and two Gaussians at 510.8 and 539.5 keV, respectively. This resulted in a value of $S_{540} = (319 \pm 56)$ for the number of events, with a fit of χ^2/n.d.f. = 0.76. In a similar manner, the number of events in the 590.8 keV peak was obtained by fitting the spectrum to the (560–625) keV energy region with the sum of the exponential with four Gaussians at 569.7, 583.2, 590.8, and 609.3 keV (χ^2/n.d.f. = 1.4):$S_{591} = (278 \pm 53)$. Thus, the peaks are observed with more than a 5σ significance level. The results of the fit are shown in Figure 1, left. Taking into account the detection efficiencies for the 539.5 keV and for the 590.8 keV γ lines (calculated by EGS4 [50] and GEANT4 [51] simulations), one obtains $T_{1/2} = 6.6^{+1.4}_{-1.0} \times 10^{20}$ year for the 539.5 keV peak and $T_{1/2} = 7.2^{+1.7}_{-1.2} \times 10^{20}$ year for the 590.8 keV peak. Combining these results, we obtain the half-life: $T_{1/2} = [6.9^{+1.0}_{-0.8}(\text{stat.}) \pm 0.7(\text{syst.})] \times 10^{20}$ year, where the systematic uncertainties are related to the uncertainty of the mass of the ^{100}MoO$_3$ sample (0.01%), the enrichment in ^{100}Mo (0.3%), and the calculation of the measurements' live time (0.5%), with a major contribution from the calculation of the efficiencies [2].

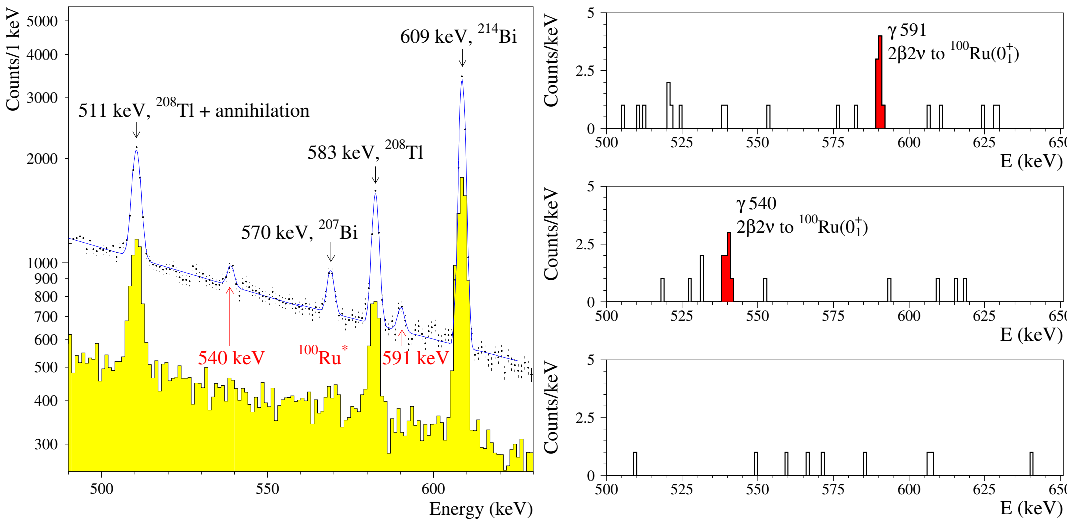

Figure 1. Left: (Color on-line) Energy spectrum collected with the ^{100}MoO$_3$ sample (points with error bars) in the (490–630) keV energy region, together with the fit (continuous curve). The background spectrum (normalized to 18120 h) is also shown (filled-in histogram). Both the 540 and 591 keV peaks of the $2\nu2\beta$ decay ^{100}Mo $\rightarrow$ ^{100}Ru(0^+_1) are clearly visible in the energy spectrum of the ^{100}MoO$_3$ sample. **Right**: (Color on-line) The coincidence energy spectra accumulated over a period of 17807 h with the ^{100}MoO$_3$ sample in the four-HPGe setup when the energy of one detector was fixed at the value expected for the ^{100}Mo $\rightarrow$ ^{100}Ru(0^+_1) $2\nu2\beta$ decay: (540 ± 2) keV (top) and (591 ± 2) keV (middle). The bottom figure shows the background obtained by shifting the energy of one detector to (545 ± 2) keV.

The two-dimensional energy spectrum of the events with multiplicity 2, accumulated in coincidence mode over a period of 17807 h, was also analyzed. Fixing the energy of one detector to the expected energy of a certain γ enables the observation of coincident signals in the other detectors with energies that correspond to γs emitted in cascade with the first one. By fixing the energy of one of the detectors to the expected energy of the γs emitted in the $2\nu2\beta$ decay of ^{100}Mo to ^{100}Ru(0^+_1) (540 or 591 keV; width of the window: ± 2 keV, in accordance with the energy resolution of the HPGe at these energies), the coincidence peak at the corresponding supplemental energy

(591 or 540 keV) is observed. These coincidence spectra are shown in Figure 1, right. The bottom part of the figure shows the background events when the energy window is shifted to the neighboring value, (545 ± 2) keV. Taking into account the efficiency calculated for the 540 keV and 591 keV γs in cascade (8.0×10^{-4} with GEANT4 [51]), the eight events detected in coincidence correspond to a half-life of $T_{1/2} = 6.8^{+3.7}_{-1.8} \times 10^{20}$ year for the $2\nu2\beta$ decay of ^{100}Mo $\rightarrow$ ^{100}Ru(0^+_1). This value is in agreement with the half-life derived from the one-dimensional spectrum ($6.9^{+1.2}_{-1.1} \times 10^{20}$ year). However, it has a much larger statistical uncertainty because of the small number of measurements (only eight events).

The data collected deep underground at the LNGS by the ARMONIA experiment allowed the observation of the $2\nu2\beta$ decay of ^{100}Mo to the 0^+_1 excited level of ^{100}Ru ($E_{exc} = 1130.3$ keV). The half-life values derived from the two-dimensional experimental spectrum of the coincidence events and from the one-dimensional spectrum are in perfect agreement. This observation does not confirm the negative result [47]; on the other hand, the measured half-life values are in agreement with the results of previous experiments [38,41–43,46].

3. Search for Double Beta Decay in ^{116}Cd with the AURORA Experiment

The ^{116}Cd isotope is one of the best candidates to search for the $0\nu2\beta$ occurrence owing to the high Q-value of $Q_{2\beta} = 2813.49(13)$ keV [34], the relatively large isotopic abundance of $\delta = 7.512(54)\%$ [33], the possibility of enrichment by ultra-centrifugation in large amounts, and the promising estimations of the decay probability [52–55]. A new search for double-beta processes in ^{116}Cd was carried out by the AURORA experiment with two ^{116}CdWO$_4$ crystal scintillators (580 g and 582 g) enriched in ^{116}Cd to 82% [56,57]. Good optical and scintillation properties of the detectors were obtained due to the high purification of ^{116}Cd and W and to the advantage of the low-thermal-gradient Czochralski technique used to grow the crystals. The active source approach (high detection efficiency), the low levels of internal contamination in U, Th, and K, and the possibility of α/β pulse shape discrimination (PSD) were exploited to reach the best sensitivities to date in the search for several 2β decay modes of ^{116}Cd.

In the AURORA experiment [3], the two ^{116}CdWO$_4$ crystals were installed in the low-background DAMA/R&D setup at LNGS. The scintillators were fixed inside polytetrafluoroethylene containers filled with ultrapure liquid scintillator and viewed through low-radioactive quartz light-guides by two 3-inch low-radioactivity photomultiplier tubes (PMTs) (Hamamatsu R6233MOD, Hamamatsu, Japan). To reduce the external background, the passive shield was made of high-purity copper (10 cm), low-radioactivity lead (15 cm), cadmium (1.5 mm), and polyethylene/paraffin (4–10 cm). In order to remove environmental radon, the setup was enclosed inside a Plexiglas box continuously flushed by high-purity nitrogen gas. An event-by-event DAQ system based on a 1 GS/s 8-bit transient digitizer (Acqiris DC270, Plan-les-Ouates, Switzerland) recorded the amplitude, the arrival time, and the pulse shape of the events. The energy scale and resolution of the detector were checked periodically with ^{22}Na, ^{60}Co, ^{137}Cs, ^{133}Ba, and ^{228}Th sources. The energy resolution of the ^{116}CdWO$_4$ detector for 2615 keV quanta of ^{208}Tl was an FWHM of $\approx 6\%$.

The pulse profiles of the events were analyzed by using the optimal filter method [58,59] to discriminate $\gamma(\beta)$ from α events. Thus, the PSD was applied to reduce the background and to estimate, by means of a time–amplitude analysis [60], the ^{228}Th contamination of the ^{116}CdWO$_4$ crystals. In order to reject the fast decay chain, ^{212}Bi $\rightarrow$^{212}Po, from the ^{232}Th family, a front-edge analysis was also performed. The ^{116}CdWO$_4$ crystal scintillators are highly radiopure, with 0.020(1) mBq/kg of ^{228}Th, $<$0.006 mBq/kg of ^{226}Ra, and 0.22(9) mBq/kg of ^{40}K, and the total U/Th α activity is 2.14(2) mBq/kg.

The energy spectrum of $\gamma(\beta)$ events from the data, collected over 26831 h with the ^{116}CdWO$_4$ detectors, is shown in Figure 2, left. It was fitted in the (660–3300) keV energy region by the model built from the $2\nu2\beta$ decay of ^{116}Cd; the internal contamination by ^{40}K, ^{232}Th, and ^{238}U; and the contribution from external γs. The model functions were simulated by the Monte Carlo code with the EGS4 package [50], and the initial kinematics of the particles emitted in the decays were given by the DECAY0 event generator [61]. The fit results in $T_{1/2} = 2.63^{+0.11}_{-0.12} \times 10^{19}$ year for the half-life of ^{116}Cd relative to the $2\nu2\beta$ decay to the ground state of ^{116}Sn; this result gives the highest accuracy to date for the half-life measurement of the $2\nu2\beta$ decay of ^{116}Cd (with a signal-to-background ratio of $\simeq$2.6 in the (1.1–2.8) MeV energy interval).

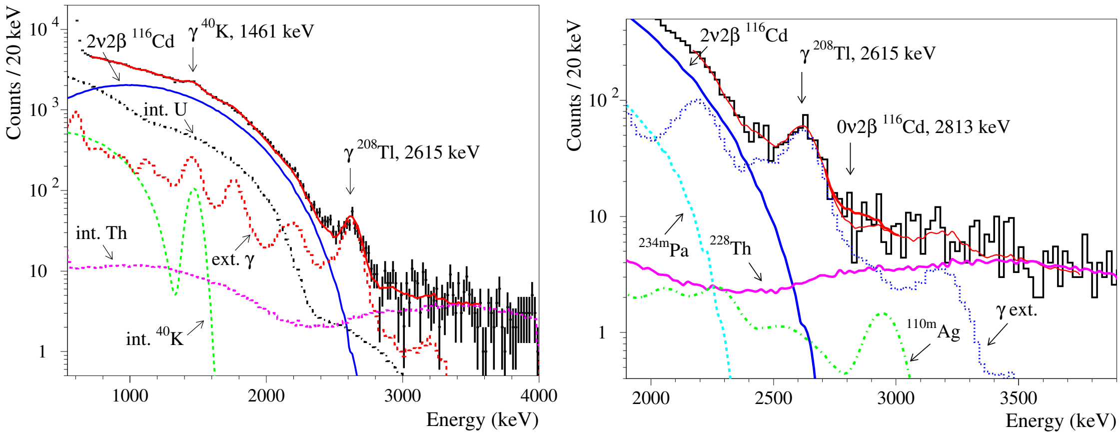

Figure 2. Energy spectrum of $\gamma(\beta)$ events collected by the ^{116}CdWO$_4$ detectors in the region of interest for $2\nu2\beta$ decay (on the left, T = 26831 h) and $0\nu2\beta$ decay (on the right, T = 35324 h) of ^{116}Cd. Also shown are the main components of the background model: the $2\nu2\beta$ decay of ^{116}Cd, the internal contamination of the ^{116}CdWO$_4$ crystals by U/Th, K ("int. U", "int. Th", "^{40}K") and contributions from external γs ("ext. γ" or "ext. Th."). The peak of the $0\nu2\beta$ decay of ^{116}Cd excluded at 90% C.L. is also shown.

To derive a limit on the ^{116}Cd $0\nu2\beta$ decay, we also included in the analysis the data from the previous stage of the experiment with a similar background rate in the region of interest (ROI): $\approx$0.1 counts/keV/kg/year. In the (2.5–3.2) MeV energy interval, the measured energy spectrum was approximated by the background model built from the distributions of the $0\nu2\beta$ (effect searched for) and $2\nu2\beta$ decays of ^{116}Cd, the internal contamination of the crystals by ^{228}Th, and the contribution from external γs (mainly from the thorium contamination in the surrounding materials). The energy resolution at the $Q_{2\beta}$ was extrapolated from calibrations with standard γ sources and is equal to an FWHM of $\approx$170 keV; for details, see Reference [57]. The fit gives an area of the expected peak of $S = (-4.5 \pm 14.2)$ counts, which means there is no evidence of the effect. In accordance with Reference [62], 19.1 counts can be excluded at 90% C.L., which leads to a new limit on the $0\nu2\beta$ decay of ^{116}Cd to the ground state of ^{116}Sn: $T_{1/2} > 2.2 \times 10^{23}$ year. The half-life limit corresponds to the effective Majorana neutrino mass limit $\langle m_\nu \rangle < (1.0$–$1.7)$ eV, obtained by using the recent nuclear matrix elements reported in References [52–55], the phase space factor from Reference [63], and the value of the axial-vector coupling constant $g_A = 1.27$. New improved limits on other 2β processes in ^{116}Cd (decays with Majoron emission, transitions to excited levels of ^{116}Sn) were set at a level of $T_{1/2} > (3.6$–$6.3) \times 10^{22}$ year.

4. Search for Double Beta Decay in ^{106}Cd with the DAMA/CRYS Setup

The experimental sensitivities for the search for double beta-plus processes (double electron capture 2ε, electron capture with positron emission $\varepsilon\beta^+$, and emission of two positrons $2\beta^+$) are substantially more modest with respect to $2\beta^-$ processes, and only indications exist for the allowed $2\nu2\varepsilon$ mode in ^{130}Ba [64,65] and ^{78}Kr [66,67] with the half-lives between 10^{20} and 10^{22} year.

One should note that a strong motivation to search for neutrinoless 2ε and $\varepsilon\beta^+$ decays is related to the possibility of refining the mechanism of the $0\nu2\beta^-$ decay: either it appears because of the neutrino Majorana mass or because of the contribution of right-handed admixtures in weak interactions [68].

The ^{106}Cd isotope is a very interesting nucleus in which to search for double beta-plus processes because of its high-energy release during decay, $Q_{2\beta} = 2775.39(10)$ keV [34], and a relatively high natural isotopic abundance of $\delta = 1.245(22)\%$ [33]. Moreover, it is also favored for possible resonant $0\nu2\varepsilon$ transitions to excited levels of ^{106}Pd [69,70]. Thus, ^{106}Cd is one of the most investigated nuclei [69].

A new experiment to search for double beta decay in ^{106}Cd is being conducted in the DAMA/CRYS setup at LNGS using a ^{106}CdWO$_4$ crystal scintillator (215 g) that is enriched in ^{106}Cd to 66%. This is the third stage of DAMA experimentation with this crystal scintillator. In the first stage, in the low-background DAMA/R&D setup, the ^{106}CdWO$_4$ crystal was fixed inside a cavity filled with high-purity silicon oil and viewed by two low-radioactivity PMTs through ∼20 cm long light-guides. A sensitivity of $T_{1/2} \sim (10^{20}–10^{21})$ year was reached for different channels of the double beta decay of ^{106}Cd [69]. In the second stage of the experiment, the ^{106}CdWO$_4$ crystal was viewed by a low-radioactivity PMT through a (archaeological) lead tungstate (^{arch}PbWO$_4$) crystal light-guide. It was installed in the central well of the ultralow-background GeMulti setup in the STELLA facility at LNGS. Limits on the 2ε, $\varepsilon\beta^+$, and $2\beta^+$ processes in ^{106}Cd were slightly improved [71] in comparison with the first stage [69].

The presently running experiment is being realized to increase the detection efficiencies of the coincidence events; thus, the ^{106}CdWO$_4$ was installed in coincidence with two large-volume low-background CdWO$_4$ crystal scintillators in close geometry. A scheme of the setup is given in Figure 3.

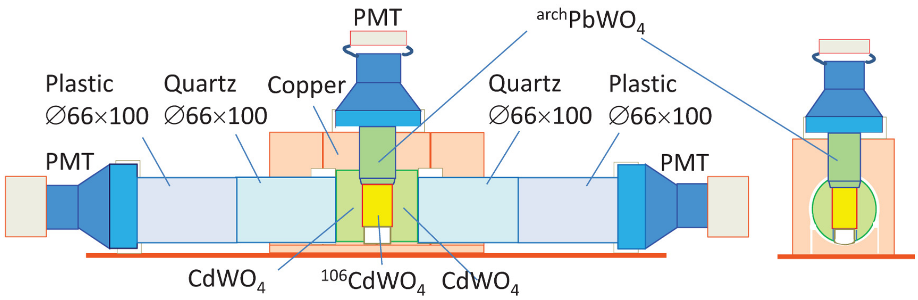

Figure 3. Schematic view of the ^{106}CdWO$_4$ setup that is now running in the DAMA/CRYS setup at LNGS.

The ^{106}CdWO$_4$ crystal scintillator is in a vertical position, as viewed through a ^{arch}PbWO$_4$ crystal light-guide by a low-radioactivity PMT (Hamamatsu R6233MOD). The ^{arch}PbWO$_4$ was developed from highly purified [72] archaeological lead [73]. The ^{106}CdWO$_4$ is almost entirely enclosed by two shaped CdWO$_4$ crystal scintillators, which are coupled to two low-radioactivity EMI9265–B53/FL PMTs through light-guides made by high-purity quartz and polystyrene. A copper structure maintains the detectors in a fixed position and also acts as a shield; the system was installed in the low-background DAMA/CRYS setup, which consists of a passive shield made of high-purity copper (11 cm), lead (10 cm), cadmium (2 mm), and polyethylene (10 cm). Moreover, to protect the detectors from environmental air, the setup is sealed and continuously flushed by high-purity nitrogen gas. The amplitude, the arrival time, and the pulse shape of the events are recorded by an event-by-event data acquisition system equipped with a 100 MSamples/s, 14-bit transient digitizer (DT5724 by CAEN, Viareggio, Italy) over a time window of 60 µs The β decay of ^{113}Cd and ^{113m}Cd, which is not of interest for this measurement, dominate the low-energy part of the ^{106}CdWO$_4$ spectrum; thus, to considerably reduce the stored data, the scintillation events of ^{106}CdWO$_4$ with an energy $\leq$ 500 keV are recorded by the DAQ only if there is a coincidence signal in at least one of the two CdWO$_4$ crystal scintillators.

The measurements started in May 2016 and are still in progress. The ^{106}CdWO$_4$ and two large CdWO$_4$ scintillators are calibrated with ^{22}Na, ^{60}Co, ^{133}Ba, ^{137}Cs, and ^{228}Th γ sources. To discriminate $\gamma(\beta)$ events from those induced by α particles, the difference in the pulse shapes in the CdWO$_4$ scintillators can be used. A preliminary data set was investigated in order to evaluate the PSD capability of the detectors in the present configuration by using various pulse shape analyses. Presently, the separation of the α and γ populations is worse than that obtained in the first stage of the experiment [69]; further analyses are in progress.

A preliminary time–amplitude analysis was performed on the data collected over 6935 h; in this way [60,74], by studying the arrival time and the energy of each event, it is possible to tag the fast α decay chain in the ^{232}Th family: ^{224}Ra (Q_α = 5.79 MeV, $T_{1/2}$ = 3.66 d) $\rightarrow$ ^{220}Rn (Q_α = 6.41 MeV, $T_{1/2}$ = 55.6 s) $\rightarrow$ ^{216}Po (Q_α = 6.91 MeV, $T_{1/2}$ = 0.145 s) $\rightarrow$ ^{212}Pb. To select α events in the decay chain, the quenching of the scintillation output in the $CdWO_4$ scintillator was considered (the so-called α/β ratio, i.e., the ratio between the α peak position in the γ-calibrated scale of a detector and the energy of the alpha particles). From this preliminary analysis, the contamination of ^{228}Th in the $^{106}CdWO_4$ crystal was estimated to be 5(1) µBq/kg.

Considering that, for some decay modes, the detection efficiencies (evaluated by Monte Carlo simulations) for coincidence events in the region of interest are 4–5 times larger with respect to the previous stage of the experiment, one can expect an improved experimental sensitivity to be obtained for the half-lives of some decay modes of ^{106}Cd to be in the range of (10^{20}–10^{22}) year; this will allow us to explore the two-neutrino $\epsilon\beta^+$ decay mode in the range of some theoretical predictions.

5. Preliminary Results for ^{150}Nd 2β Decay with the GeMulti Setup

The high-energy release $Q_{2\beta}$ = 3371.38(20) keV [34] and the high natural isotopic abundance δ = 5.638(28)% [33] highlight the ^{150}Nd nuclide as one of the most promising 2β decaying isotope among the 35 naturally occurring ones [32]. The ^{150}Nd $2\nu2\beta$ decay to the ground state of ^{150}Sm was measured in several direct experiments to be in the range $T_{1/2}$ = (0.7–1.9) $\times$ 10^{19} year [75–77]. In addition, the transition to the first excited level of ^{150}Sm was observed with a half-life in the range of $T_{1/2}$ = (7–14)$\times10^{19}$ year [78–80].

In this new measurement, a sample of high-purity Nd_2O_3 (total mass of 2.381 kg), compressed into 20 cylindrical tablets ((56 $\pm$ 1) mm in diameter with a (16 $\pm$ 0.5) mm thickness), was installed in the GeMulti ultralow-background HPGe gamma-spectrometer (see Section 2). The energy scale and resolution of the HPGe detectors were measured at the beginning of the experiment with γ-sources. Then, the four spectra were equalized to the same energy scale by using background gamma peaks. As a result, the gamma peak positions in the cumulative spectrum deviate by less than 0.2 keV from the table values.

The radioactive contamination of the Nd_2O_3 sample before and after the applied purification process was measured as reported in [4]. In particular, the Nd_2O_3 sample was contaminated by ^{138}La and ^{176}Lu. The two-dimensional energy spectrum of coincidences between two detectors (events with multiplicity 2), accumulated over 16375 h with the Nd_2O_3 sample, was analyzed. The 2β decay of ^{150}Nd to the first 0^+_1 excited level of ^{150}Sm is followed by the emission of γs in cascade with energies of 334.0 keV and 406.5 keV, respectively. By fixing the energy of the events in one of the detectors to the energy of the γ expected to be emitted in a cascade after the 2β decay of ^{150}Nd to the first 0^+_1 excited level of ^{150}Sm, a signal with energy corresponding to the other γs in cascade is expected. Fixing the energy of one of the detectors to the expected energy with the energy window ±1.4 $\times$ FWHM, the coincidence signals at the supplemental energy (406.5 or 334.0 keV, respectively) were observed (see Figure 4).

The area of each peak was estimated and, taking into account the detection efficiency, the half-life of the 2β decay ^{150}Nd $\rightarrow$ ^{150}Sm (0^+_1, 740.5 keV) was preliminarily determined as $T_{1/2} = 4.7^{+4.1}_{-1.9} \times 10^{19}$ year. This half-life is in agreement with the results of the previous experiments (see Reference [4] and references therein). The experiment is presently running to enhance the statistics in order to improve the half-life value accuracy.

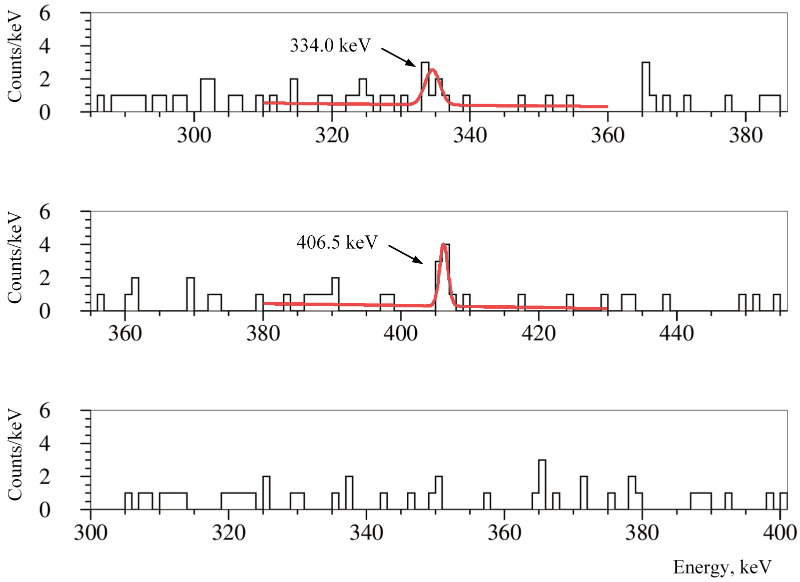

Figure 4. Coincidence energy spectra measured by the GeMulti setup with the 2.381 kg Nd_2O_3 sample over 16,375 h when the energy in one detector was fixed to the energy interval at which γs from the ^{150}Nd $\rightarrow$ ^{150}Sm (0^+_1, 740.5 keV) decay—406.5 keV $\pm$ 1.4 $\times$ FWHM (top), 334.0 keV $\pm$ 1.4 $\times$ FWHM (middle)—are expected. The bottom spectrum shows a random coincidence background in the energy range of interest when the energy of events in one of the detectors is taken as 375 keV $\pm$ 1.4 $\times$ FWHM (no γs with this energy are expected in either the ^{150}Nd 2β decay nor in the decays of nuclides that are radioactive contaminants of the Nd_2O_3 sample or of the setup).

6. Conclusions

In this report, the main results obtained with DAMA experimental setups in the search for rare processes and double beta decay are briefly summarized. Some further details are given about the main results of ARMONIA and AURORA experiments. Finally, a summary is provided of the status of (1) the new measurements of ^{106}Cd 2β decay using a $^{106}CdWO_4$ detector and (2) the study of the $2\nu2\beta$ decay of ^{150}Nd to the first 0^+_1 excited level of ^{150}Sm using a Nd_2O_3 sample in the GeMulti HPGe γ setup. Data collection is in progress, and the study of further purification procedures for the samples of various compounds containing interesting isotopes for the purpose of establishing further improved sensitivities is ongoing.

Author Contributions: All the authors of this paper have been significantly contributing to the presented results working on the various aspects of the different phases of this experiment.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Bernabei, R.; Belli, P.; Bussolotti, A.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Dai, C.J.; d'Angelo, A.; Di Marco, A.; He, H.L.; et al. First model independent results from DAMA/LIBRA–phase2. *Universe* **2018**, *4*, 116. [CrossRef]
2. Belli, P.; Bernabei, R.; Boiko, R.S.; Cappella, F.; Cerulli, R.; Danevich, F.A.; d'Angelo, S.; Incicchitti, A.; Kobychev, V.V.; Kropivyansky, B.N.; et al. New observation of $2\beta2\nu$ decay of ^{100}Mo to the 0^+_1 level of ^{100}Ru in the ARMONIA experiment. *Nucl. Phys. A* **2010**, *846*, 143–156. [CrossRef]
3. Polischuk, O.G.; Barabash, A.S.; Belli, P.; Bernabei, R.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Chernyak, D.M.; Danevich, F.A.; d'Angelo, S.; et al. Investigation of 2β decay of ^{116}Cd with the help of enriched $^{116}CdWO_4$ crystal scintillators. *AIP Conf. Proc.* **2017**, *1894*, 020018. [CrossRef]

4. Barabash, A.S.; Belli, P.; Bernabei, R.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Danevich, F.A.; Di Marco, A.; Incicchitti, A.; Kasperovych, R.V.; et al. Double beta decay of ^{150}Nd to the first excited 0^{+} level of ^{150}Sm: Preliminary results. *Nucl. Phys. At. Energy* **2018**, *19*, 95–102. [CrossRef]
5. Belli, P.; Bernabei, R.; Cappella, F.; Cerulli, R.; Dai, C.J.; Danevich, F.A.; d'Angelo, A.; Incicchitti, A.; Kobychev, V.V.; Nagorny, S.S.; et al. Search for α decay of natural Europium. *Nucl. Phys. A* **2007**, *789*, 15–29. [CrossRef]
6. Belli, P.; Bernabei, R.; Cappella, F.; Cerulli, R.; Danevich, F.A.; Incicchitti, A.; Laubenstein, M.; Nagorny, S.S.; Nisi, S.; Polischuk, O.G.; et al. First observation of α decay of ^{190}Pt to the first excited level ($E_{exc} = 137.2$ keV) of ^{186}Os. *Phys. Rev. C* **2011**, *83*, 034603. [CrossRef]
7. Belli, P.; Bernabei, R.; Bukilic, N.; Cappella, F.; Cerulli, R.; Dai, C.J.; Danevich, F.A.; de Laeter, J.R.; Incicchitti, A.; Kobychev, V.V.; et al. Investigation of β decay of ^{113}Cd. *Phys. Rev. C* **2007**, *76*, 064603. [CrossRef]
8. Bernabei, R.; Belli, P.; Cappella, F.; Cerulli, R.; Montecchia, F.; Nozzoli, F.; Incicchitti, A.; Prosperi, D.; Tretyak, V.I.; Zdesenko, Y.G.; et al. Search for β and $\beta\beta$ decays in ^{48}Ca. *Nucl. Phys. A* **2002**, *705*, 29–39. [CrossRef]
9. Bernabei, R.; Belli, P.; Cappella, F.; Caracciolo, V.; Castellano, S.; Cerulli, R.; Dai, C.J.; d'Angelo, A.; Di Marco, A.; He, H.L.; et al. New search for correlated e^{+} e^{-} pairs in the α decay of ^{241}Am. *Eur. Phys. J. A* **2013**, *49*, 64. [CrossRef]
10. Bernabei, R.; Belli, P.; Cerulli, R.; Montecchia, F.; Nozzoli, F.; Incicchitti, A.; Prosperi, D.; Dai, C.J.; He, H.L.; Kuang, H.H.; et al. Search for solar axions by Primakoff effect in NaI crystals. *Phys. Lett. B* **2001**, *515*, 6–12. [CrossRef]
11. Belli, P.; Bernabei, R.; Cappella, F.; Cerulli, R.; Danevich, F.A.; d'Angelo, A.; Incicchitti, A.; Kobychev, V.V.; Laubenstein, M.; Polischuk, O.G.; et al. Search for ^{7}Li solar axions using resonant absorption in LiF crystal: Final results. *Phys. Lett. B* **2012**, *711*, 41–45. [CrossRef]
12. Belli, P.; Bernabei, R.; Cerulli, R.; Danevich, F.A.; d'Angelo, A.; Goriletski, V.I.; Grinvov, B.V.; Incicchitti, A.; Kobychev, V.V.; Laubenstein, M.; et al. ^{7}Li solar axions: Preliminary results and feasibility studies. *Nucl. Phys. A* **2008**, *806*, 388–397. [CrossRef]
13. Cappella, F.; Cerulli, R.; Incicchitti, A. A preliminary search for Q-balls by delayed coincidences in NaI(Tl). *Eur. Phys. J. direct C* **2002**, *14*, 1–6. [CrossRef]
14. Bernabei, R.; Belli, P.; Cerulli, R.; Montecchia, F.; Amato, M.; Ignesti, G.; Icicchitti, A.; Prosperi, D.; Dai, C.J.; He, H.L.; et al. Extended Limits on Neutral Strongly Interacting Massive Particles and Nuclearites from NaI(Tl) Scintillators. *Phys. Rev. Lett.* **1999**, *83*, 4918. [CrossRef]
15. Bernabei, R.; Belli, P.; Cappella, F.; Cerulli, R.; d'Angelo, A.; Emiliani, F.; Incicchitti, A. Search for Daemons with NEMESIS. *Mod. Phys. Lett. A* **2012**, *27*, 1250031. [CrossRef]
16. Belli, P.; Bernabei, R.; Dai, C.J.; He, H.L.; Ignesti, G.; Icicchitti, A.; Kuang, H.H.; Ma, J.M.; Montecchia, F.; Ponkratenko, O.A.; et al. New experimental limit on the electron stability and non-paulian transitions in Iodine atoms. *Phys. Lett. B* **1999**, *460*, 236–241. [CrossRef]
17. Belli, P.; Bernabei, R.; Di Nicolantonio, W.; Landoni, V.; Incicchitti, A.; Prosperi, D.; Dai, C.J.; Bacci, C. Charge conservation and electron lifetime: Limits from a liquid xenon scintillator. *Astrop. Phys.* **1996**, *5*, 217–219. [CrossRef]
18. Belli, P.; Bernabei, R.; Dai, C.J.; Ignesti, G.; Icicchitti, A.; Montecchia, F.; Ponkratenko, O.A.; Prosperi, D.; Tretyak, V.I.; Zdesenko, Y.G. Quest for electron decay $e^{-} \to \nu_e \gamma$ with a liquid xenon scintillator. *Phys. Rev. D* **2000**, *61*, 117301. [CrossRef]
19. Bernabei, R.; Belli, P.; Cappella, F.; Montecchia, F.; Nozzoli, F.; d'Angelo, A.; Incicchitti, A.; Prosperi, D.; Cerulli, R.; Dai, C.J.; et al. Search for spontaneous transition of nuclei to a superdense state. *Eur. Phys. J. A* **2005**, *23*, 7–10. [CrossRef]
20. Bernabei, R.; Belli, P.; Cappella, F.; Montecchia, F.; Nozzoli, F.; d'Angelo, A.; Incicchitti, A.; Prosperi, D.; Cerulli, R.; Dai, C.J.; et al. A search for spontaneous emission of heavy clusters in the ^{127}I nuclide. *Eur. Phys. J. A* **2005**, *24*, 51–56. [CrossRef]
21. Bernabei, R.; Belli, P.; Montecchia, F.; Nozzoli, F.; d'Angelo, A.; Cappella, F.; Incicchitti, A.; Prosperi, D.; Castellano, S.; Cerulli, R.; et al. Performances and potentialities of a $LaCl_3$: Ce scintillator. *Nucl. Instr. Meth. A* **2005**, *555*, 270–281. [CrossRef]

22. Bernabei, R.; Amato, M.; Belli, P.; Cerulli, R.; Dai, C.J.; Denisov, V.Y.; He, H.L.; Incicchitti, A.; Kuang, H.H.; Ma, J.M.; et al. Search for the nucleon and di-nucleon decay into invisible channels. *Phys. Lett. B* **2000**, *493*, 12–18. [CrossRef]
23. Bernabei, R.; Belli, P.; Montecchia, F.; Nozzoli, F.; Cappella, F.; Incicchitti, A.; Prosperi, D.; Cerulli, R.; Dai, C.J.; Denisov, V.Y.; et al. Search for rare processes with DAMA/LXe experiment at Gran Sasso. *Eur. Phys. J. A* **2006**, *27*, 35–41. [CrossRef]
24. Bernabei, R.; Belli, P.; Cappella, F.; Cerulli, R.; Dai, C.J.; d'Angelo, A.; d'Angelo, S.; Di Marco, A.; He, H.L.; Incicchitti, A.; et al. Search for charge non-conserving processes in ^{127}I by coincidence technique. *Eur. Phys. J. C* **2012**, *72*, 1920. [CrossRef]
25. Bernabei, R.; Belli, P.; Montecchia, F.; Nozzoli, F.; d'Angelo, A.; Capella, F.; Incicchitti, A.; Prosperi, D.; Castellano, S.; Cerulli, R.; et al. Search for possible charge non-conserving decay of ^{139}La into ^{139}Ce with $LaCl_3$(Ce) scintillator. *Ukr. J. Phys.* **2006**, *51*, 1037–1043.
26. Belli, P.; Bernabei, R.; Dai, C.J.; He, H.L.; Ignesti, G.; Incicchitti, A.; Kuang, H.H.; Ma, J.M.; Montecchia, F.; Ponkratenko, O.A.; et al. New limits on the nuclear levels excitation of ^{127}I and ^{23}Na during charge nonconservation. *Phys. Rev. C* **1999**, *60*, 065501. [CrossRef]
27. Belli, P.; Bernabei, R.; Dai, C.J.; Ignesti, G.; Incicchitti, A.; Montecchia, F.; Ponkratenko, O.A.; Prosperi, D.; Tretyak, V.I.; Zdesenko, Y.G. Charge non-conservation restrictions from the nuclear levels excitation of ^{129}Xe induced by the electron's decay on the atomic shell. *Phys. Lett. B* **1999**, *465*, 315–322. [CrossRef]
28. Bernabei, R.; Belli, P.; Cappella, F.; Cerulli, R.; Dai, C.J.; d'Angelo, A.; He, H.L.; Incicchitti, A.; Kuang, H.H.; Ma, J.M.; et al. New search for processes violating the Pauli exclusion principle in sodium and in iodine. *Eur. Phys. J. C* **2009**, *62*, 327–332. [CrossRef]
29. Bernabei, R.; Belli, P.; Montecchia, F.; de Sanctis, M.; di Nicolantonio, W.; Incicchitti, A.; Prosperi, D.; Bacci, C.; Dai, C. J.; Ding, L.K.; et al. Search for non-paulian transitions in ^{23}Na and ^{127}I. *Phys. Lett. B* **1997**, *408*, 439–444. [CrossRef]
30. Belli, P.; Bernabei, R.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Danevich, F.A.; Di Marco, A.; Incicchitti, A.; Poda, D.V.; Polischuk, O.G.; et al. Investigation of rare nuclear decays with BaF_2 crystal scintillator contaminated by radium. *Eur. Phys. J. A* **2014**, *50*, 134. [CrossRef]
31. Belli, P.; Bernabei, R.; Cappella, F.; Cerulli, R.; Danevich, F.A.; Denisov, V.Y; d'Angelo, A.; Incicchitti, A.; Kobychev, V.V.; Poda, D.V.; et al. Search for long-lived superheavy eka-tungsten with radiopure $ZnWO_4$ crystal scintillator. *Phys. Scr.* **2015**, *90*, 085301. [CrossRef]
32. Tretyak, V.I.; Zdesenko, V.I. Tables of double beta decay data-an update. *Atom. Data Nucl. Data* **2002**, *80*, 83–116. [CrossRef]
33. Meija, J.; Coplen, T.B.; Berglund M.; Brand, W.A.; De Bièvre, P.; Gröning, M.; Holden, N.E.; Irrgeher, J.; Loss, R.D.; Walczyk, T.; et al. Isotopic compositions of the elements 2013 (IUPAC Technical Report). *Pure Appl. Chem.* **2016**, *88*, 293–306. [CrossRef]
34. Wang, M.; Audi, G.; Kondev, F.G.; Huang, W.J.; Naimi, S.; Xu, X. The AME2016 atomic mass evaluation (II). Tables, graphs and references. *Chin. Phys. C* **2017**, *41*, 030003. [CrossRef]
35. Hidaka, H.; Ly, C.V.; Suzuki, M. Geochemical evidence of the double β decay of ^{100}Mo. *Phys. Rev. C* **2004**, *70*, 025501. [CrossRef]
36. Barabash, A.S. Average and recommended half-life values for two-neutrino double beta decay. *Nucl. Phys. A* **2015**, *935*, 52–64. [CrossRef]
37. Armengaud, E.; Augier, C.; Barabash, A.S.; Beeman, J.W.; Bekker, T.B.; Bellini, F.; Benoît, A.; Bergé, L.; Bergmann, T.; Billard, J.; et al. Development of ^{100}Mo-containing scintillating bolometers for a high-sensitivity neutrinoless double-beta decay search. *Eur. Phys. J. C* **2017**, *77*, 785. [CrossRef]
38. Barabash, A.S.; Avignone, F.T., III; Collar, J.I.; Guerard, C.K.; Arthur, R.J.; Brodzinski, R.L.; Miley, H.S.; Reeves, J.H.; Meier, J.R.; Ruddick, K.; et al. Two neutrino double-beta decay of ^{100}Mo to the first excited 0^+ state in ^{100}Ru. *Phys. Lett. B* **1995**, *345*, 408–413. [CrossRef]
39. Barabash, A.S.; Avignone, F.T., III; Guerard, C.K.; Brodzinski, R.L.; Miley, H.S.; Reeves, J.H.; Umatov, V.I. Proceedings of the 26th Rencontre de Moriond: Festschrift Wuthrick (JP)-11th Moriond Workshop Massive Neutrinos Test of Fundamental Symmetries. 1991; p. 77. Available online: https://cds.cern.ch/record/227770/files/C91-01-26_Proceedings.pdf (accessed on 12 December 2018).

40. Barabash, A.S.; Avignone, F.T., III; Guerard, C.K.; Brodzinski, R.L.; Miley, H.S.; Reeves, J.H.; Umatov, V.I. Two neutrino double-beta decay of ^{100}Mo to the first excited 0^+ state in ^{100}Ru. In Proceedings of the 3rd International Symposium WEIN'92, Dubna, Russia, 16–22 June 1992; World Scientific: Singapore, 1993; p. 582.
41. Barabash, A.S. Gurriaran, R.; Hubert, F.; Hubert, P.; Umatov, V.I. $2\nu\beta\beta$ decay of ^{100}Mo to the first 0^+ excited state in ^{100}Ru. *Phys. At. Nucl.* **1999**, *62*, 2039–2043.
42. Arnold, R.; Augier, C.; Baker, J.; Barabash, A.S.; Bongrand, M.; Broudin, G.; Brudanin, V.; Caffrey, A.J.; Egorov, V.; Etienvre, A.I.; et al. Measurement of double beta decay of ^{100}Mo to excited states in the NEMO 3 experiment. *Nucl. Phys. A* **2007**, *781*, 209–226. [CrossRef]
43. Kidd, M.F.; Esterline, J.H.; Tornow, W.; Barabash, A.S.; Umatov, V.I. New results for double-beta decay of ^{100}Mo to excited final states of ^{100}Ru using the TUNL-ITEP apparatus. *Nucl. Phys. A* **2009**, *821*, 251–261. [CrossRef]
44. De Braeckeleer, L.; Hornish, M.; Barabash, A.S.; Umatov, V.I. Measurement of the $\beta\beta$-Decay Rate of ^{100}Mo to the First Excited 0^+ State of ^{100}Ru. *Phys. Rev. Lett.* **2001**, *86*, 3510. [CrossRef] [PubMed]
45. Hornish, M.J.; De Braeckeleer, L.; Barabash, A.S.; Umatov, V.I. Double β decay of ^{100}Mo to excited final states. *Phys. Rev. C* **2006**, *74*, 044314. [CrossRef]
46. Arnold, R.; Augier, C.; Barabash, A.S.; Basharina-Freshville, A.; Blondel, S.; Blot, S.; Bongrand, M.; Brudanin, V.; Busto, J.; Caffrey, A.J.; et al. Investigation of double beta decay of ^{100}Mo to excited states of ^{100}Ru. *Nucl. Phys. A* **2014**, *925*, 25–36. [CrossRef]
47. Blum, D.; Bust, J.; Campagne, J.E.; Dassié, D.; Hubert, F.; Hubert, P.; Isaac, M.C.; Izac, C.; Jullian, S.; Kouts, B.N.; et al. Search for γ-rays following $\beta\beta$ decay of ^{100}Mo to excited states of ^{100}Ru. *Phys. Lett. B* **1992**, *275*, 506–511. [CrossRef]
48. Belli, P.; Bernabei, R.; Boiko, R.S.; Cerulli, R.; Danevich, F.A.; d'Angelo, S.; Incicchitti, A.; Kobychev, V.V.; Kropivyansky, B.N.; Laubenstein, M.; et al. Preliminary results on the search for ^{100}Mo 2β decay to the first excited 0^+_1 level of ^{100}Ru. In Proceedings of the International Conference "Current Problems in Nuclear Physics and Atomic Energy", Kyiv, Ukraine, 29 May–3 June 2006; pp. 479–482.
49. Belli, P.; Bernabei, R; Boiko, R.S.; Cappella, F.; Cerulli, R.; Danevich, F. A.; d'Angelo, S.; Incicchitti, A.; Kobychev, V.V. ; Kropivyansky, B.N.; et al. Preliminary results on the search for ^{100}Mo 2β decay to the first excited 0^+_1 level of ^{100}Ru (ARMONIA Experiment). In Proceedings of the International Conference Current Problems in Nuclear Physics and Atomic Energy, Kyiv, Ukraine, 29 May–3 June 2006; pp. 473–476.
50. Nelson, W.R.; Hirayama, H.; Rogers, D.W.O. *The EGS4 CODE SYSTEM*; Technical report SLAC-265; Stanford Linear Accelerator Center Stanford University: Stanford, CA, USA, 1985.
51. Agostinelli, S.; Allison, J.; Amako, K.; Apostolakis, J.; Araujo, H.; Arce, P.; Asai, M.; Axen, D.; Banerjee, S.; Barrand, G.; et al. Geant4-a simulation toolkit. *Nucl. Instrum. Meth. A* **2003**, *506*, 250–303. [CrossRef]
52. Rodryguez, T.R.; Martynez-Pinedo, G. Energy Density Functional Study of Nuclear Matrix Elements for Neutrinoless $\beta\beta$ Decay. *Phys. Rev. Lett.* **2010**, *105*, 252503. [CrossRef]
53. Simkovic, F.; Rodin, V.; Faessler, A.; Vogel, P. $0\nu\beta\beta$ and $2\nu\beta\beta$ nuclear matrix elements, quasiparticle random-phase approximation, and isospin symmetry restoration. *Phys. Rev. C* **2013**, *87*, 045501. [CrossRef]
54. Hyvarinen, J.; Suhonen, J. Nuclear matrix elements for $0\nu\beta\beta$ decays with light or heavy Majorana-neutrino exchange. *Phys. Rev. C* **2015**, *91*, 024613. [CrossRef]
55. Barea, J.; Kotila, J.; Iachello, F. $0\nu\beta\beta$ and $2\nu\beta\beta$ nuclear matrix elements in the interacting boson model with isospin restoration. *Phys. Rev. C* **2015**, *91*, 034304. [CrossRef]
56. Barabash, A.S.; Belli, P.; Bernabei, R.; Boiko, R.S.; Cappella, F.; Caracciolo, V.; Chernyak, D.M.; Cerulli, R.; Danevich, F.A.; Di Vacri, M.L.; et al. Low background detector with enriched $^{116}CdWO_4$ crystal scintillators to search for double β decay of ^{116}Cd. *JINST* **2011**, *6*, P08011. [CrossRef]
57. Barabash, A.S.; Belli, P.; Bernabei, R.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Chernyak, D.M.; Danevich, F.A.; d'Angelo, S.; Incicchitti, A.; et al. Final results of the Aurora experiment to study 2β decay of ^{116}Cd with enriched $^{116}CdWO_4$ crystal scintillators. *Phys. Rev. D* **2018**, *98*, 092007. [CrossRef]
58. Gatti, E.; De Martini, F. A new linear method of discrimination between elementary particles in scintillation counters. In *Nuclear Electronics II, Proceedings of the Conference on Nuclear Electronics, V. II, Belgrade, Yugoslavia, 15–20 May 1961*; Brüder Rosenbaum: Vienna, Austria, 1962; pp. 265–276.

59. Bardelli, L.; Bini, M.; Bizzeti, P.G.; Carraresi, L.; Danevich, F.A.; Fazzini, T.F.; Grinyov, B.V.; Ivannikova, N.V.; Kobychev, V.V.; Kropivyansky, B.N.; et al. Further study of $CdWO_4$ crystal scintillators as detectors for high sensitivity 2β experiments: Scintillation properties and pulse-shape discrimination. *Nucl. Instr. Meth. A* **2006**, *569*, 743–753. [CrossRef]
60. Danevich, F.A.; Kobychev, V.V.; Ponkratenko, O.A.; Tretyak, V.I.; Zdesenko, Y.G. Quest for double beta decay of ^{160}Gd and Ce isotopes. *Nucl. Phys. A* **2001**, *694*, 375–391. [CrossRef]
61. Ponkratenko, O.A.; Tretyak, V.I.; Zdesenko, Y.G. Event generator DECAY4 for simulating double-beta processes and decays of radioactive nuclei. *Phys. At. Nucl.* **2000**, *63*. [CrossRef]
62. Feldman, G.J.; Cousins, R.D. Unified approach to the classical statistical analysis of small signals. *Phys. Rev. D* **1998**, *57*, 3873. [CrossRef]
63. Kotila, J.; Iachello, F. Phase-space factors for double-β decay. *Phys. Rev. C* **2012**, *85*, 034316. [CrossRef]
64. Meshik, A.P.; Hohenberg, C.M.; Pravdivtseva, O.V.; Kapusta, Y.S. Weak decay of ^{130}Ba and ^{132}Ba: Geochemical measurements. *Phys. Rev. C* **2001**, *64*, 035205. [CrossRef]
65. Pujol, M.; Marty, B.; Burnard, P.; Philippot, P. Xenon in Archean barite: Weak decay of ^{130}Ba, mass-dependent isotopic fractionation and implication for barite formation. *Geochim. Cosmochim. Acta* **2009**, *73*, 6834–6846. [CrossRef]
66. Gavrilyuk, Y.M.; Gangapshev, A.M.; Kazalov, V.V.; Kuzminov, V.V.; Panasenko, S.I.; Ratkevich, S.S. Indications of $2\nu2K$ capture in ^{78}Kr. *Phys. Rev. C* **2013**, *87*, 035501. [CrossRef]
67. Ratkevich, S.S.; Gangapshev, A.M.; Gavrilyuk, Y.M.; Karpeshin, F.F.; Kazalov, V.V.; Kuzminov, V.V.; Panasenko, S.I.; Trzhaskovskaya, M.B.; Yakimenko, S.P. Comparative study of the double-K-shell-vacancy production in single- and double-electron-capture decay. *Phys. Rev. C* **2017**, *96*, 065502. [CrossRef]
68. Hirsch, M.; Muto, K.; Oda, T.; Klapdor-Kleingrothaus, H.V. Nuclear structure calculation of $\beta^+\beta^+$, $\beta^+/$EC and EC/EC decay matrix elements. *Z. Phys. A* **1994**, *347*, 151–160. [CrossRef]
69. Belli, P.; Bernabei, R.; Boiko, R.S.; Brudanin, V.B.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Chernyak, D.M.; Danevich, F.A.; d'Angelo, S.; et al. Search for double-β decay processes in ^{106}Cd with the help of a $^{106}CdWO_4$ crystal scintillator. *Phys. Rev. C* **2012**, *85*, 044610. [CrossRef]
70. Krivoruchenko, M.I.; Šimkovic, F.; Frekerse, D.; Faessler, A. Resonance enhancement of neutrinoless double electron capture. *Nucl. Phys. A* **2011**, *859*, 140–171. [CrossRef]
71. Belli, P.; Bernabei, R.; Brudanin, V.B.; Cappella, F.; Caracciolo, V.; Cerulli, R.; Chernyak, D.M.; Danevich, F.A.; d'Angelo, S.; Di Marco, A.; et al. Search for 2β decay of ^{106}Cd with an enriched $^{106}CdWO_4$ crystal scintillator in coincidence with four HPGe detectors. *Phys. Rev. C* **2016**, *93*, 045502. [CrossRef]
72. Boiko, R.S.; Virich, V.D.; Danevich, F.A.; Dovbush, T.I.; Kovtun, G.P.; Nagornyi, S.S.; Nisi, S.; Samchuk, A.I.; Solopikhin, D.A.; Shcherban', A.P. Ultrapurification of archaeological lead. *Inorg. Mater.* **2011**, *47*, 645–648. [CrossRef]
73. Danevich, F.A.; Kim, S.K.; Kim, H.J.; Kim, Y.D.; Kobychev, V.V.; Kostezh, A.B.; Kropivyansky, B.N.; Laubenstein, M.; Mokina, V.M.; Nagorny, S.S.; et al. Ancient Greek lead findings in Ukraine. *Nucl. Instr. Meth. A* **2009**, *603*, 328–332. [CrossRef]
74. Danevich, F.A.; Georgadze, A.S.; Kobychev, V.V.; Kropivyansky, B.N.; Kuts, V.N.; Nikolaiko, A.S.; Tretyak, V.I.; Zdesenko, Y. The research of 2β decay of ^{116}Cd with enriched $^{116}CdWO_4$ crystal scintillators. *Phys. Lett. B* **1995**, *344*, 72–78. [CrossRef]
75. Artemiev, V.; Brakchmana, E.; Karelina, K.; Kirichenko, V.; Klimenko, A.; Kozodaeva, O.; Lubimov, A.; Mitin, A.; Osetrov, S.; Paramokhin, V.; et al. Half-life measurement of ^{150}Nd $2\beta2\nu$ decay in the time projection chamber experiment. *Phys. Lett. B* **1995**, *345*, 564–568. [CrossRef]
76. De Silva, A.; Moe, M.K.; Nelson, M.A.; Vient, M.A. Double β decays of ^{100}Mo and ^{150}Nd. *Phys. Rev. C* **1997**, *56*, 2541. [CrossRef]
77. Arnold, R.; Augier, C.; Baker, J.D.; Barabash, A.S.; Basharina-Freshville, A.; Blondel, S.; Blot, S.; Bongrand, M.; Brudanin, V.; Busto, J.; et al. The NEMO-3 Collaboration Measurement of the $2\nu\beta\beta$ decay half-life of ^{150}Nd and a search for $0\nu\beta\beta$ decay processes with the full exposure from the NEMO-3 detector. *Phys. Rev. D* **2016**, *94*, 072003. [CrossRef]
78. Barabash, A.S.; Hubert, F.; Hubert, P.; Umatov, V.I. Double beta decay of ^{150}Nd to the first 0^+ excited state of ^{150}Sm. *JETP Lett.* **2004**, *79*, 10–12. [CrossRef]

79. Barabash, A.S.; Hubert, P.; Nachab, A.; Umatov, V.I. Investigation of $\beta\beta$ decay in ^{150}Nd and ^{148}Nd to the excited states of daughter nuclei. *Phys. Rev. C* **2009**, *79*, 045501. [CrossRef]
80. Kidd, M.F.; Esterline, J.H.; Finch, S.W.; Tornow, W. Two-neutrino double-β decay of ^{150}Nd to excited final states in ^{150}Sm. *Phys. Rev. C* **2014**, *90*, 055501. [CrossRef]