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FIRST RESULT OF THE EXPERIMENTAL SEARCH FOR THE $2K$-CAPTURE OF ^{124}Xe WITH THE COPPER PROPORTIONAL COUNTER

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The first result of experiment on searching for $2K$-capture of ^{124}Xe with the large-volume copper proportional counter (LPC) is given. The 12-liter sample with 63.3% (44 g) of ^{124}Xe was used in measurements. The limit on the half-life of ^{124}Xe with regard to $2K(2\nu)$-capture for the ground state of ^{124}Te ($0^+ \to 0^+$, g.s.) has been found: $T_{1/2} \geqslant 4.67 \cdot 10^{20}$ y (90% C.L.). A sample with the volume of 52 liters comprising ^{124}Xe (10.6 l — 58.6 g) and ^{126}Xe (14.1 l — 79.3 g) will be used at the next step of the experiment to increase a sensitivity of $2K$-capture of ^{124}Xe registration. In this case, sensitivity to the investigated process will be at the level of $S = 1.46 \cdot 10^{21}$ y (90% C.L.) for 1 year measurement.

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INTRODUCTION

The Baksan Neutrino Observatory INR RAS has several unique low background laboratories. One of them is deep underground low-background laboratory (DULB-4900) [1]. The laboratory is located at a distance of 3700 m from the main entry to the Baksan Neutrino Observatory tunnel, in the hall of $\sim 6 \times 6 \times 40$ m. Thickness of the mountain rock over DULB corresponds to 4900 m.w.e., thereby decreasing cosmic ray flux by $\sim 10^7$ times. These allow one to carry out experiments on searching for rare process and decay. An experiment on searching for $2K$-capture of ^{124}Xe is an example.

Theoretical calculations for $2K(2\nu)$ within the frames of different models (QRPA, MCM) predict the following half-life periods with respect to $2K$-capture of ^{124}Xe: $1.08 \cdot 10^{22}$ y [2], $3.9 \cdot 10^{23}$ y [3]. The values were obtained by taking the fraction of $2K(2\nu)$-capture events in ^{124}Xe with respect to the total number of $2e(2\nu)$-capture events to be 73% [4].

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1. BASIC ASSUMPTIONS

When two electrons are captured from the K-shell in ^{124}Xe, a daughter atom of ^{124}Te** is formed with two vacancies in the K-shell. The technique to search for this reaction is based on the assumption that the energies of characteristic photons and the probability that they will be emitted when a double vacancy is filled, are the same as the sum of respective values when two single vacancies of the K-shell in two singly ionized Te* atoms are filled. In such a case, the total measured energy is $2K_{ab} = 63.62$ keV, where K_{ab} is the binding energy of a K electron in a Te atom (27.47 keV). The fluorescence yield upon filling of a single vacancy in the K-shell of Te is 0.857. The energies and relative intensities of the characteristic lines in the K series are $K_{\alpha 1} = 27.47$ keV (100%), $K_{\alpha 2} = 27.20$ keV (54%), $K_{\beta 1} = 30.99$ keV (18%), and $K_{\beta 2} = 31.7$ keV (5%) [5]. There are three possible ways for de-excitation of a doubly ionized K-shell: 1) emission of Auger electrons only (e_a, e_a); 2) emission of a single characteristic quantum and an Auger electron (K, e_a); and 3) emission of two characteristic quanta and low-energy Auger electrons (K, K, e_a), with probabilities of $p_1 = 0.020$, $p_2 = 0.246$, and $p_3 = 0.734$, respectively. A characteristic quantum can travel a distance long enough in a gas medium between the points of its production and absorption. But photoelectrons produce almost pointwise charge clusters of primary ionization in the gas. In the case of the event with the escape of two characteristic quanta absorbed in the working gas and a single Auger electron, the energy is distributed among three pointwise charge clusters. It is these three-point (or three-cluster) events possessing a unique set of features that were the subject of the search in [6].

To register the process of $2K$-capture in ^{124}Xe, a large proportional counter (LPC) with a casing of M1-grade copper has been used [7]. The LPC is inside the shielding of 18 cm thick copper, 15 cm thick lead, and 8 cm thick borated polyethylene layers. The installation is placed in one of the chambers of the underground laboratory DULB-4900, where cosmic ray flux is lowered to the level of $(3.03 \pm 0.10) \cdot 10^{-9}$ cm$^{-2} \cdot$ s^{-1} [8].

The details of the counter signals formation, of the features of pulses registration by digital oscilloscope, of the data treatment and analysis are described in [7, 9, 10].

2. MEASUREMENT RESULTS

The measurements with radioactive-pure xenon (background sample) have been done at the initial stage of the experiment for understanding background level of experimental setup and for future comprising with ^{124}Xe. The LPC pressure is equal to 1.9 atm. The measurement time was 970 h. Count rate in the interval 15–150 keV was found to be ~ 16 h^{-1}.

Table 1. **Characteristics of samples**

Samples, l	Content, l								
	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
No. 1a, 1.98	1.02	3.81e-1	5.19e-1	3.1e-4	1.3e-4	3.76e-5	1.8e-5	1.8e-5	1.8e-5
No. 2, 1.27	1.13	1.3e-2	3.4e-2	6.1e-4	1.2e-4	3.6e-5	3.6e-5	3.5e-5	3.5e-5
No. 3, 3.56	1.60	1.93	1.03e-3	6.8e-5	6.8e-5	6.7e-5	6.7e-5	6.6e-5	6.5e-5
No. 4, 3.11	1.50	1.4	1.2e-1	6.7e-4	6.7e-4	5.8e-4	5.8e-4	5.8e-4	5.8e-4
No. 5, 1.32	1.11	4.0e-3	7.5e-5	5.0e-5	5.0e-5	3.1e-4	5.0e-5	5.0e-5	5.0e-5
No. 6, 1.31	1.31	1.6e-4	1.3e-5	1.3e-5	1.3e-5	1.3e-5	1.2e-5	1.2e-5	1.2e-5

A sample of 12 l of xenon enriched in the isotope ^{124}Xe to 63.3% (44 g) was used in the main measurements with ^{124}Xe. This sample combines several ones, see Table 1.

The LPC was filled to the maximum pressure $P_{\max} = 1.1$ atm and the first test measurement was done during 310 h. Count rate in the interval 15–150 keV was found to be ~ 60 h^{-1}. Excess background was created by ^{85}Kr decays. This isotope is a part of natural krypton (~ 50 ppm) which is dissolved in xenon.

To reduce a background of krypton, an attempt was made to "wash out" the ^{124}Xe sample by "clean krypton" without ^{85}Kr. This procedure is based on the assumption that xenon will settle on a bottle wall at the liquid nitrogen temperature ($-190\,°$C) (the pressure of saturated vapor $1.93 \cdot 10^{-3}$ mm Hg) but krypton will stay in gas phase because it has a relatively large value of saturated vapor pressure (~ 1.8 mm Hg). It was planned preliminarly to add into the xenon a few cm^3 of the krypton and then to repeat this procedure several times. The rate of xenon treating was controlled by residual background of LPC. Unfortunately, this procedure did not give any positive results. One can assume that all krypton has been absorbed by xenon similar to a charcoal trap.

A helium was chosen as another "washing" gas. The 0.5 l of a helium was added to the bottle with the xenon per one procedure. The bottle has been frozen and helium was exhausted. The background of LPC in the interval 15–150 keV became ~ 12 h^{-1}, i.e., the desired result was achieved after six procedures. Easy and chip procedure for gas cleaning from remnant of krypton was developed as a result of this work. A comparison of spectra of ^{124}Xe sample before purification (spectrum *1*), of the radioactive-pure xenon (spectrum *2*) and of ^{124}Xe after purification (spectrum *3*) are shown in Fig. 1. All spectra are normalized to 100 h.

The LPC was filled to the maximum possible for xenon sample pressure — 1.1 atm to perform the main measurements on search for $2K$-capture of ^{124}Xe. The efficiency of registration of two K photons with the energy of ~ 27 keV is equal to 0.09 at this pressure. Measurements on the underground low-background setup were carried out during 1130 h. The charge pulses are recorded by digital

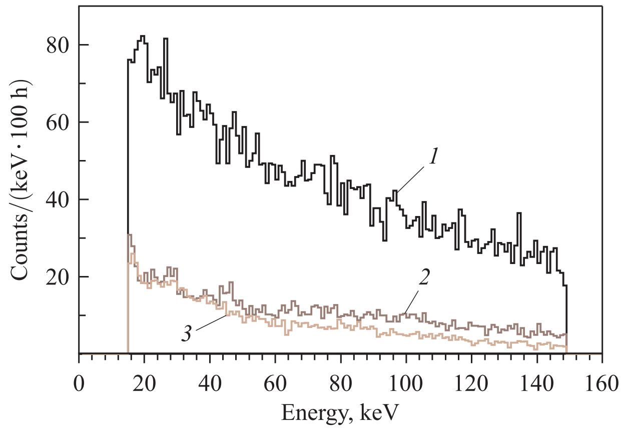

Fig. 1. The spectra of xenon before purification (*1* — ^{124}Xe + ^{85}Kr sample, 1.1 atm), radioactive-pure xenon (*2* — $^{\rm nat}$Xe sample, 1.9 atm), and xenon after purification (*3* — ^{124}Xe sample, 1.1 atm) normalized to 100 h

oscilloscope to the personal computer. The collected dataset was treated by special program. As a result, the entire dataset was divided into groups (spectra): one-, two-, and three-point events. The full spectrum, the spectra of one-, two-, and three-point events are shown in Fig. 2, *a* by the black line, the gray line, the gray bar graph, and the black bar graph. The three-point events spectrum is shown in Fig. 2, *b* more precisely. The "useful" effect is searched for in the three-point event spectrum in the energy interval (63.6 ± 3.7) keV. The amplitudes of partial pulses for each event are arranged in the increased order $[(A_1, A_2, A_3) \to (m_0 \leqslant m_1 \leqslant m_2)]$, to simplify the selection of events with a given set of features. Selected useful events must satisfy the selection criteria $5.0 \leqslant m_0 \leqslant 13.0$ keV and $m_1/m_2 \geqslant 0.7$. The events corresponding to these ratios were selected from the spectrum of the three-point pulses to reduce the background. They are presented in Fig. 2, *c*.

The area of the expected effect is marked by dashed lines. There are not remained any events in this area after the event selection. Therefore, we obtain $N_{\rm eff} = 2.44$ for 1130 h ($\mu = 0.00{-}2.44$) or $N_{\rm eff} = 21.37$ y^{-1} (90% C.L.) by using the recommendation of the work [11] for the given (effect+background) value (0 events). The half-life period has been calculated using the formula:

$$\lim T_{1/2} \geqslant \ln 2N \frac{p_3 \varepsilon_p \varepsilon_3}{N_{\rm eff}},$$

where $N = 2.59 \cdot 10^{23}$ is the number of ^{124}Xe atoms in the operating volume of the counter, $p_3 = 0.735$ is a portion of $2K$-captures accompanied by the emission

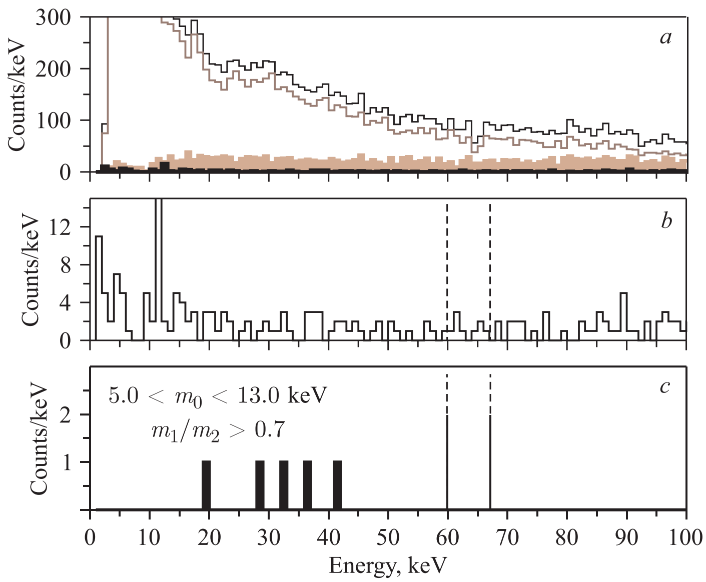

Fig. 2. Amplitude spectra of LPC, 1.1 atm of xenon enriched in the isotope ^{124}Xe to 63.3% (44 g): *a*) the black line — all events; the gray line — one-point events; the gray bar graph — two-point events; the black bar graph — three-point events; *b*) the three-point event spectra — full; *c*) the three-point event spectra — selected by criteria $5.0 \leqslant m_0 \leqslant 13.0$ keV and $m_1/m_2 \geqslant 0.7$. The pure measurement time is 1130 h. The dashed lines indicate the boundaries of the region of interest — (63.6 ± 3.7) keV

of two quanta; $\varepsilon_p = 0.09$ is the probability of two K-quanta absorption in the operating volume; $\varepsilon_3 = 0.422$ is the selection efficiency for three-point events due to $2K$-capture in ^{124}Xe.

The result obtained is

$$T_{1/2}(0\nu + 2\nu, 2K) \geqslant 4.6 \cdot 10^{20} \text{ y (90\% C.L.)}.$$

It should be noted that for the "normal" working pressure of 5 atm, efficiency of registration of two K-quanta is equal to 0.809. A sensitivity of experimental setup to the required process at this value of efficiency is equal to

$$S = 1.46 \cdot 10^{21} \text{ y (90\% C.L.)}.$$

A new 58 l sample No. 7 of the enriched xenon was obtained by the team at the last time. The characteristics of the sample are shown in Table 2.

The LPC was filled to 5 atm by new xenon sample No. 7 for checking background measurement. The spectrum (*1*) of sample No. 7 background obtained

Table 2. **Characteristics of samples No. 7, total volume** ~ 58 **l,** ^{124}Xe $\sim$ (4.33 **l** $\approx$ 23.96 **g),** ^{126}Xe $\sim$ (15.24 **l** $\approx$ 85.7 **g)**

Samples, l	Content, l								
	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
No. 7, 58	4.33	15.24	24.147	14.033	0.0511	0.0394	0.0168	0.0606	0.0543

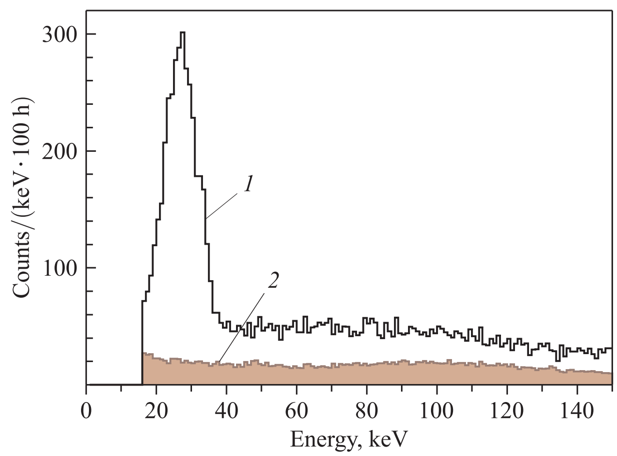

Fig. 3. Amplitude spectra of LPC with xenon sample No. 7 (5 atm) (spectrum *1*) and radioactive-pure xenon (5 atm) (spectrum *2*)

at 146 h is shown in Fig. 3. The spectrum (*2*) of the radioactive-pure xenon is shown for comparison also. All spectra are normalized to 100 h. It is seen from the spectra comparison that sample No. 7 has radioactive impurities. It creates a background in a wide energy range including the region of interest. Intense peak is visible in area of 28 keV. Differences of the spectra (*1*, *2*) give a count of rate ~ 48 h^{-1} in the interval 15–150 keV. It turned out that this value decreases exponentially over time with a half-life of about 30 days. The radioactive isotope ^{127}Xe was identified as the source due to the set of the attributes. The decay scheme of isotope ^{127}Xe [12] is shown in Fig. 4. The isotope ^{127}Xe can be produced by secondary cosmic rays in isotopes of sample No. 7 through reactions $^{126}\mathrm{Xe}(n,\gamma)^{127}$Xe and $^{128}\mathrm{Xe}(\mu,n)^{127}$Xe. There are no published data on the cross sections of the reactions mentioned above. Apparently, this can be explained by the fact that in a natural xenon, content of the original isotopes is extremely small — ^{126}Xe — 0.09%, ^{128}Xe — 1.91%. Sample No. 7 is highly enriched in these isotopes, and it can be used to measure the previously mentioned cross sections.

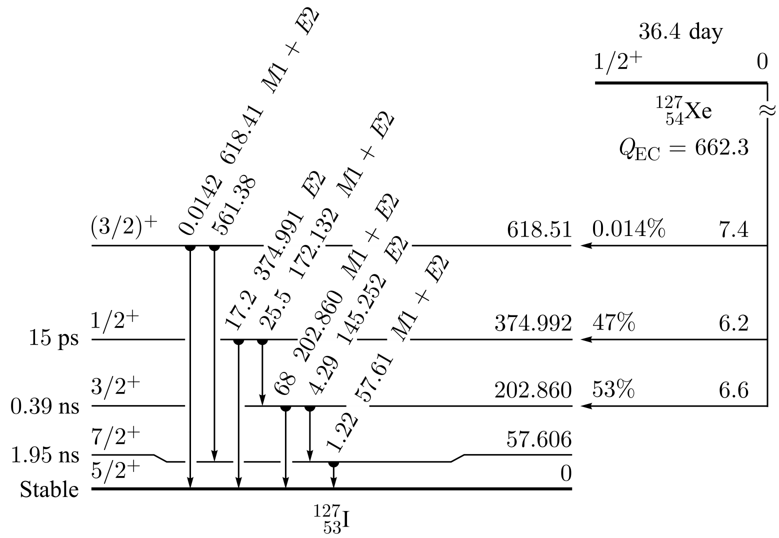

Fig. 4. The decay scheme of isotope ^{127}Xe [12]

The time of about 4 months is required to decrease the ^{127}Xe background to acceptable level before using of xenon sample No. 7 in the experiment on searching for $2K$-capture of ^{124}Xe. A significant amount of ^{126}Xe is present in the sample No. 7. This isotope can also decay by $2K$-capture. The transition energy is 897 keV. A high limit for the $2K$-capture half-life of ^{124}Xe could be obtained simultaneously if sample No. 7 will be used in experiment on searching for the $2K$-capture of ^{124}Xe.

CONCLUSIONS

The new experiment on searching for $2K$-capture of ^{124}Xe is done. The 12 l sample with 63.3% (44 g) of ^{124}Xe was used in measurements. The limit on the half-life of ^{124}Xe with regard to $2K(2\nu)$-capture for the ground state of ^{124}Te ($0^+ \rightarrow 0^+$, g.s.) has been found: $T_{1/2} \geqslant 4.67 \cdot 10^{20}$ y (90% C.L.).

A new 52 l sample of enriched xenon will be used on the next stage of the experiment. This sample has as part of ^{124}Xe (10.6 l — 58.6 g) and ^{126}Xe (14.1 l — 79.3 g). A sensitivity at 1-year measurement time of the experimental setup to the required process will be equal to

$$S = 1.46 \cdot 10^{21} \text{ y } (90\% \text{ C.L.}).$$

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