Rare decays of mercury nuclei

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In the Solotvina Underground Laboratory an experiment has been carried out (1108.6 hours) in which an ultralow-background HP Ge detector of volume 165 cm^3 was used to establish the following limits on the probability of rare decays of mercury nuclei: $0\nu 2K$-capture of ^{196}Hg—$T_{1/2} \geqslant 9.6 \cdot 10^{17}$ years, $(0\nu + 2\nu)2K$-capture of ^{196}Hg—$T_{1/2} \geqslant 2.5 \cdot 10^{18}$ years, transition of mercury nuclei to a superdense state—$\tau \geqslant 10^{22}$ years, decay with emission of ^{24}Ne or ^{28}Mg—$T_{1/2} \geqslant (1.3\text{–}3.7) \cdot 10^{21}$ years.

In preparation of an experiment on search for 2β decay of ^{76}Ge in the Solotvina Underground Laboratory of the Nuclear Research Institute of the Ukrainian Academy of Sciences, which is located in a salt mine at a depth of 1000 m.w.e.,[1] measurements were made for 1108.6 hours of the background of a semiconductor detector made from ultrapure germanium (HP Ge).[2]

In an installation with passive and active shielding we used a detector made by the West German firm PGT with an active volume 165 cm^3 (relative efficiency 38.7%) and an energy resolution of 1.9 keV for the 1.33-MeV line. The detector was repackaged in a low-background cryostat made of titanium and oxygen-free copper. The first layer of passive shielding of the semiconductor detector is mercury, which is in titanium containers and practically completely surrounds the detector. Inside the cryostat there is also a volume of mercury which protects the crystal from penetration of background radiation through the gap between the cold finger and the walls of the opening in the outer mercury shield. The thickness of the mercury layer varied in different directions from 10 cm to 30 cm. Then there are layers of oxygen-free copper (11–15 cm), lead (23–30 cm), and polyethylene (24 cm). The active shield of plastic scintillator ($117 \times 114 \times 9$ cm^3) serves to further suppress the muon component of cosmic rays.

As a result the integral background of the semiconductor detector in the region 100–2850 keV is 20.6 counts per hour, and the background counting rate in the energy region of the $0\nu 2\beta$ decay of ^{76}Ge is 2.4 counts/year·keV·kg, which corresponds to the level of the best low-background installations with HP-Ge detectors. This circumstance, and also the fact that directly around the detector there is 570 kg of mercury, permitted establishment in the present work of limits on the probability of the following processes: $2K$ capture of ^{196}Hg, transition of mercury nuclei into a superdense state, and decay of mercury of nuclei with emission of clusters.

2*K* CAPTURE OF ^{196}Hg

The theory of $2e$ capture has been developed in Refs. 3 and 4, whose main conclusions are as follows.

Double electron capture is a process in which a nucleus tears out two electrons from the atomic shell, as a result of which the charge of the nucleus decreases by two units as the result of the conversion of two protons into neutrons. In addition to radiation of two neutrinos, $2e$ capture is characterized by emission of x rays and (or) electrons which arise in filling of the vacancies in the electron shells of the daughter atom.

If we assume that the neutrino which arises in conversion of one proton into a neutron can be absorbed in capture of a second electron, then neutrinoless $2e$ capture becomes possible.

For accomplishment of this process it is necessary that lepton charge is not conserved, and that the neutrino be a Majorana particle with incomplete polarization, the cause of which may be the neutrino mass and (or) the presence in the weak interaction of an admixture of right-handed currents.

Neutrinoless $2e$ capture is considered as a two-step process. In the first step exchange of a neutrino occurs, which is accomplished as the result of the unconserved leptonic charge of the weak interaction and leads to mixing of the initial and excited daughter atomic states. In the second step the atom is de-excited by emission of x rays and (or) electrons, and the nucleus is de-excited by emission of a γ ray (or electron) which carries away the entire energy of the reaction or the difference between it and the energy of some one of the excited levels of the daughter nucleus. Detection of such γ rays with the "incorrect" energy is characteristic only of the neutrinoless process, whereas γ transitions between levels of the daughter nucleus can occur also in $2\nu 2e$ capture to one of these levels.

One of the objects for the search for $2e$ decay is the nucleus ^{196}Hg. Level schemes of ^{196}Hg, ^{196}Au, and ^{196}Pt are given in Fig. 1.[5] The relative abundance of ^{196}Hg is 0.15%, and the mass difference of the atoms of ^{196}Hg and ^{196}Pt is 820 ± 3 keV.[6] When the binding energy of the K electrons is taken into account, the excitation energy available in $0\nu 2K$-capture of ^{196}Hg (663.2 ± 3 keV) can be carried away by two γ rays with energies 307.5 and 355.7 keV.

The experiment was carried out in the period January to March of 1989. The investigated portion of the background spectrum (in 1108.6 hours) is shown in Fig. 2. The γ lines which are present in it belong to radionuclides of the ^{232}Th series and also to ^{134,137}Cs.

The peaks with energy 307.5 and 355.7 keV are not present in the spectrum, and the average background counting rate in the interval 300–400 keV is $(1.8 \pm 0.1) \cdot 10^{-2}$ counts/hour·keV.

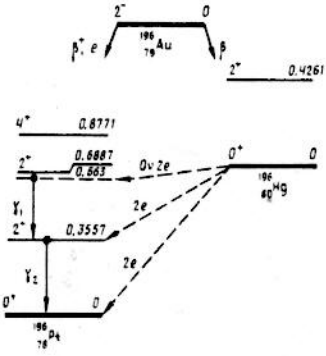

FIG. 1. Level scheme of ^{196}Hg, ^{196}Au, and ^{196}Pt ($E_{2K} = 663.2 \pm 3$ keV, $E_{\gamma 1} = 307.5 \pm 3$ keV, $E_{\gamma 2} = 355.7 \pm 0.1$ keV).

The maximum possible area of the lines being looked for which will be unnoticeable at this background level was determined by means of the following procedure. The experimental data in the vicinities of the possible peak were approximated by the method of least squares in the form of the sum of two functions: a linear background and a Gaussian (the effect) with a given location and dispersion. As a result it was found that at the 68% confidence level the area of the 307.5-keV peak does not exceed 13 counts, and that of the 355.7-keV peak does not exceed 6 counts. For conversion to half-lives of ^{196}Hg one must know the number of nuclei and the absolute efficiency of detection of these γ rays. As the result of the significant absorption in mercury of γ rays with energy 300–400 keV, in the calculations only the layer of mercury closest to the detector, of thickness 40 mm, was considered, in which there were $2.14 \cdot 10^{23}$ ^{196}Hg nuclei.

For calculation of the detection efficiency we developed a program which takes into account the geometry of the measurements, the absorption of γ rays in mercury, the container walls, and cryostat, the shape of the response function, and the intrinsic efficiency of the detector. The method of calculation was checked by comparison with the results of the calculations of Refs. 7 and 8 and also in measurements with point and volume sources of ^{137}Cs, ^{60}Co, and ^{40}K. The discrepancies between the calculation and experiment do not exceed 5%. The calculated values of the detection efficiency at the total-absorption peak of γ rays from $2K$ capture of ^{196}Hg are $6.5 \cdot 10^{-4}$ (307.5 keV) and $7.7 \cdot 10^{-4}$ (355.7 keV).

When the number of nuclei and the time of the measurements are taken into account this leads to the following estimate of the half-life of ^{196}Hg against $2K$ capture: $T_{1/2}$ (0ν channel) $\geqslant 9.6 \cdot 10^{17}$ years ($E_\gamma = 307.5$ keV) and $T_{1/2}$ ($0\nu + 2\nu$ channel) $\geqslant 2.5 \cdot 10^{18}$ years ($E_\gamma = 355.7$ keV).

These results exceed by 7 and 16 times the limits established previously for $T_{1/2}$ (respectively $1.3 \cdot 10^{17}$ years and $1.5 \cdot 10^{17}$ years).[9]

SEARCH FOR SPONTANEOUS TRANSITIONS OF MERCURY NUCLEI TO A SUPERDENSE STATE

It is well known that production of a pion condensate in nuclear matter[10–12] can lead to appearance of a second minimum in the function which expresses the dependence of the energy of the nucleus on its density. If this minimum corresponds to densities and binding energies greater than those in ordinary nuclei, the latter can undergo a spontaneous transition to a superdense state.[13,14]

For some nuclei the limiting probabilities of such transitions have been established experimentally (Table I). In Ref. 15 scintillators (sodium iodide and hexafluorobenzene) were used to detect γ rays with energy 3–10 MeV which should, according to the assumption of the authors, accompany transitions to a superdense state if the energy release exceeds 0.05–1 MeV per nucleon. In the experiment of Ref. 16 a multicrystal scintillation spectrometer sensitive to neutrons and γ rays was used. Events with multiplicity greater than three and with a total energy release in the range 8–50 MeV were detected. It was assumed that for a difference in binding energy of the order 0.5–5 MeV per nucleon the excitation of the nucleus should be removed by simultaneous emission of neutrons and γ rays.

The idea of the present work is to attempt to detect high-energy γ radiation from 570 kg of mercury ($1.72 \cdot 10^{27}$ nuclei) surrounding a HP Ge semiconductor detector, as-

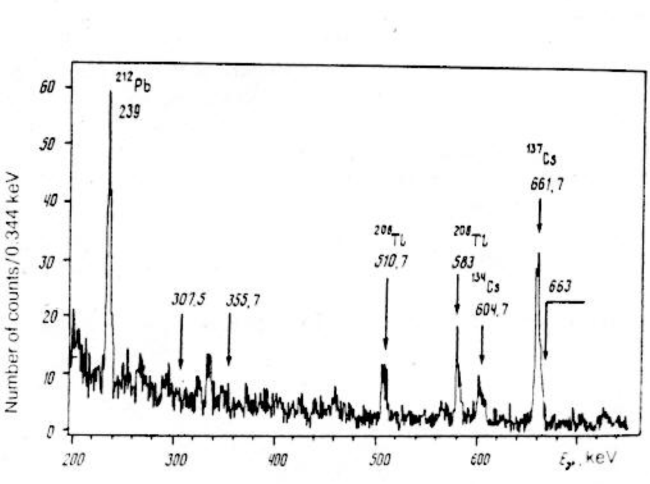

FIG. 2. Portion of the background spectrum of the high-purity Ge detector in the interval 200–750 keV.

TABLE I. Limits of half-life (in units of 10^{21} years) of various nuclei against transitions to the superdense state.

Reference	C	F	Na	Al	Cu	Cd	I	W	Pb
[15]	10	10	0,3	–	–	–	0,3	1	–
[16]	23	94	57	120	90	40	57	–	20

suming that the cause of its occurrence may be spontaneous transitions of mercury nuclei into a superdense state (as in Refs. 15 and 16 a very simple model of the mechanism of removal of excitation was adopted—the hypothetical transition is accompanied by emission of neutrons and several γ rays).

In Fig. 3 we have shown a background spectrum of the detector in the interval 0.5–26 MeV collected in 805.9 hours. In the insert we have shown a portion (3–26 MeV) in the form of a histogram with a cell width 0.5 MeV. Of the features of this spectrum we can mention the rise at an energy 5.5 MeV (14 events in the region 4–6 MeV). This rise may be due to α particles of ^{210}Po which is produced as the result of decay of ^{210}Pb (half-life 22 years) which in turn is present in natural lead. This peak in the spectrum of a low-background HP Ge detector (manufactured by PGT) was observed in Ref. 17, and its intensity decreased rapidly with reduction of the voltage applied to the semiconductor detector. As a result the authors of Ref. 17 assumed that ^{210}Po occurs in the immediate vicinity of the P contact of the Ge crystal (possibly in the gold layer), since on decrease of the voltage there is formed around the contact an insensitive zone which the ^{210}Po α particles cannot pass through (their range in germanium at this energy is 5 μm). An additional cause of background in the region 3–6 MeV may be γ rays from the (n,γ) reaction in mercury nuclei (in 100 captures 86 γ rays with energy 3–5 MeV are emitted and 40 γ rays with energy 5–6.5 MeV).

Similarly, a portion of the background counts up to energy 8 MeV can be explained by capture of neutrons in the copper and lead which serve as successive layers of the detector shielding. The upper limit of the flux of thermal neutrons is $3\cdot10^{-6}$ sec$^{-1}\cdot$cm^{-2}.[1]

In regard to events in the energy region 9–27 MeV, they may be due first of all to cosmic muons which have passed through the detector which for some reason did not result in operation of the active shield and, second, to energetic γ rays which are generated by muons in their traversal through the passive shield of the installation and also in the walls of the underground laboratory (the vertical flux of muons at the Solotvina Laboratory is $6\cdot10^{-3}$ h$^{-1}\cdot$cm^{-2}; Ref. 1).

However, the absolute background rate at energies above 4 MeV is extraordinarily low: $7.5\cdot10^{-6}$, $1.5\cdot10^{-5}$, and $(1-3)\cdot10^{-6}$ counts/h·keV for the respective intervals 4–5, 5–5.5, and 6–26 MeV. For comparison we give similar data[18] obtained by the Milan University Group in a laboratory near Mont Blanc at a depth of 5000 m.w.e. (muon flux $5\cdot10^{-6}$ h$^{-1}\cdot$cm^{-2}) with a Ge(Li) detector of volume 135 cm^3: $8.9\cdot10^{-5}$ counts/h·keV (5 MeV) and $(3-7)\cdot10^{-6}$ counts/h·keV (6–8 MeV).

As can be seen in Fig. 3, in the spectrum of the semiconductor detector there are no such peaks which could be due to detection of high-energy γ rays (total-absorption peaks and peaks from emission of one and two γ rays with energy 511 keV). As the limit of the area of such peaks we have taken the value one count, since an estimate of this quantity by means of the method of maximum likelihood or on the basis of the standard technique of least squares gives for its value less than one count (at the 95%) confidence level.

By means of the program mentioned in the previous section we calculated the total efficiency for detection by the detector of γ rays emitted by mercury nuclei. In addition, we

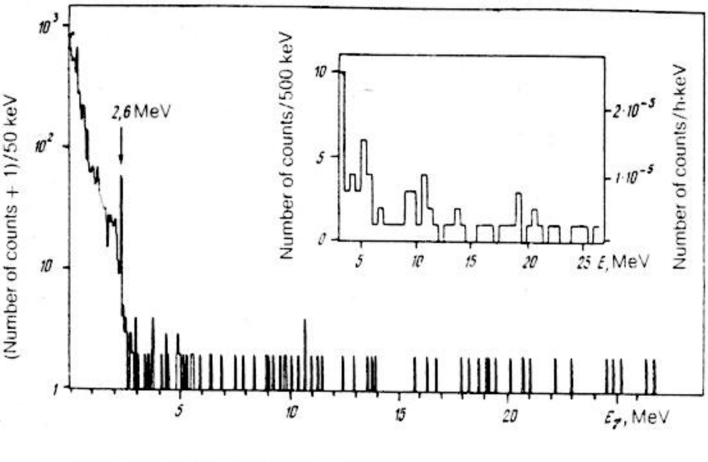

FIG. 3. Background spectrum of a high-purity Ge semiconductor detector in the region 0.5–26 MeV.

calculated the ratio of the total efficiency to the efficiency on the basis of double-escape peaks. The method of the calculation was checked against the results of Ref. 19, where the absolute efficiency was measured experimentally for detection on the basis of the photopeak and the escape peaks of a Ge(Li) semiconductor detector of volume 65 cm^3 (made by PGT) for γ rays with energy 5–16 MeV.

It was found that the total efficiency for detection of the effect on the basis of the double-escape peak with emission of the two γ rays is $7\cdot10^{-5}$ (8 MeV) and $5\cdot10^{-5}$ (16 MeV), and with emission of three γ rays it is 10^{-4} (8 MeV) and $7.5\cdot10^{-5}$ (16 MeV).

As a result we established at the 95% confidence level the following limits of the lifetime of mercury nuclei against transitions to a superdense state: $\tau \geqslant (0.8\text{–}1.5)\cdot10^{22}$ y.

Let us evaluate, proceeding from this result, the minimum height of the barrier which separates the ordinary state and the superdense state. For this purpose we shall use the theoretical dependence obtained in Ref. 20 between the barrier height U_0 and the lifetime (in units of 10^{22} years), which for mercury has the following form:

$$U_0(\text{MeV}) \geqslant 10.48\cdot(1+(4.6+\ln\tau)/55)\,[1-(n_0/n_s)^{1/3}]^{-2},$$

where n_0/n_s which is the ratio of the densities of the ordinary state and the superdense state. For $n_s = 2n_0$ the barrier height separating the ordinary state and superdense state is greater than 265 MeV, while for $n_s = 8n_0$ it is reduced to 45 MeV.

On the basis of the measurements made, limits can be obtained also on transitions to the superdense state of copper and lead nuclei, since these materials serve as the next (after mercury) layers of the detector shielding. However, calculations show that the efficiency for detection of the γ radiation from these nuclei decreases by 3–5 orders of magnitude, in view of which the lifetime limits established are substantially below those which were obtained in Ref. 16 (see Table I).

CLUSTER RADIOACTIVITY OF MERCURY NUCLEI

The discovery in 1984 of a new form of natural radioactivity—emission from ^{223}Ra nuclei of ^{14}C (Refs. 21 and 22)—produced significant interest in this phenomenon. During the five subsequent years another nine nuclides were observed which are subject to decay with emission of light ions of ^{14}C, ^{24}Ne, and ^{28}Mg,[23-25] and a theoretical interpretation was developed for cluster radioactivity, based both on analogy with α decay and on the theory of nuclear fission.[26-28] However, in spite of a satisfactory description of the experimental data, this duality of the theory leads to certain difficulties. For example, in Ref. 26 the theoretical value of the probability for ^{223}Ra lags behind the experimental value, differing by three orders of magnitude, by addition to the decay energy (the difference of the masses of the parent and daughter nuclei) of the energy of zero-point oscillations, which is essentially equivalent to violation of the conservation of energy.

On the other hand, the phenomenological systematics of the existing results follows rather well a linear dependence of the logarithm of the half-lives on the natural logarithm of the transmissions of the Coulomb barriers calculated in accordance with the simple single-particle theory of α decay.[24,25] Therefore for improvement and development of the theory it would be desirable to extend the spectrum of investigated nuclei toward both heavier and lighter nuclides. For this purpose in the present work we have established limits on the probability of cluster radioactivity of mercury nuclei.

In Table II we have listed the stable isotopes of mercury and have indicated the differences of the masses of the atoms (nuclei) in the initial and final states for decays with emission of ^{24}Ne and ^{28}Mg:[5]

$${}^{A}_{80}\text{Hg} \to {}^{24}_{10}\text{Ne} + {}^{A-24}_{70}\text{Yb},$$

$${}^{24}_{10}\text{Ne} \xrightarrow[3.38\ \text{min}]{\beta^-(1997\ \text{keV})} {}^{24}_{11}\text{Na} \xrightarrow[14.66\ \text{h}]{\beta^-(1391\ \text{keV})} {}^{24}_{12}\text{Mg};$$

$${}^{A}_{80}\text{Hg} \to {}^{28}_{12}\text{Mg} + {}^{A-28}_{68}\text{Er},$$

$${}^{28}_{12}\text{Mg} \xrightarrow[20.9\ \text{h}]{\beta^-(457\ \text{keV})} {}^{28}_{13}\text{Al} \xrightarrow[2.24\ \text{min}]{\beta^-(2863\ \text{keV})} {}^{28}_{14}\text{Si}.$$

The final result of the ^{24}Ne decay is emission of a cascade of two γ rays with energies 2754.0 and 1368.6 keV (deexcitation of the second excited level of ^{24}Mg, which results from the β^- decay of ^{24}Na). Therefore the detection in our experiment of γ rays with these energies would be an indication of the possibility of decay of mercury nuclei by this channel. In the case of ^{28}Mg the decay chain ends with radiation of a γ ray with energy 1779.0 keV (transition from the first excited level of ^{28}Si).

In the experimental spectrum the γ peaks with energies 1368.6, 1779.0, and 2754 keV are not observed. Limiting estimates were made on the basis of bands in the vicinities of 1779 and 2754 keV by means of the methods described in the previous sections. At the 95% confidence level the limit of the area of the peak is equal to one count both for 1779 keV and for 2754 keV. The detection efficiency was calculated for the entire mercury mass and amounts to $9.7\cdot10^{-5}$ (1779 keV) and $8.4\cdot10^{-5}$ (2754 keV).

TABLE II. Limits of $T_{1/2}$ (in units of 10^{21} years) established in the present work against decay of mercury nuclei with emission of ^{24}Ne and ^{28}Mg.

Nucleus	Isotopic abundance, %	Decay energy, MeV		Limit of $T_{1/2}$	
		^{24}Ne	^{28}Mg	^{24}Ne	^{28}Mg
^{198}Hg	10.1	31.92	44.16	1.3	1.5
^{199}Hg	17.0	33.93	43.18	2.2	2.5
^{200}Hg	23.1	29.92	41.98	2.9	3.4
^{201}Hg	13.2	29.26	41.10	1.7	1.9
^{202}Hg	29.7	28.29	—	3.7	—

Limits of the half-lives for the various mercury isotopes calculated for the 95% confidence level are given in the last two columns of Table II and lie in the range $(1.3\text{–}3.7)\cdot 10^{21}$ y. There are no theoretical calculations of the probability of decay of mercury nuclei with emission of ^{24}Ne and ^{28}Mg, but estimates made by us on the basis of the single-particle theory of α decay lead to half-lives which are 20–30 orders of magnitude greater than the experimental limits obtained. Nevertheless we note that the latter are the highest half-life limits established in studies of cluster radioactivity.

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