# Experimental limits on the branching ratio of double electron capture in $^{196}$Hg

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An experiment lasting 1478 h has shown that the half-life for the decay of $^{196}$Hg through neutrinoless double electron capture is, with a confidence level of 68%, longer than $1.3 \times 10^{17}$ yr. The experiment was carried out in an underground laboratory with a low-background semiconductor spectrometer.

The neutrinoless double electron capture ($0\nu 2e$ capture) is one of the $2\beta$ processes which violate lepton-charge conservation and which involve Majorana neutrinos when these particles have a mass and/or if there is an admixture of right-handed lepton currents.[1-3] These circumstances are attracting increased interest in the study of $0\nu 2e$ capture.[1-6]

A theory for $2e$ capture was derived in Refs. 1–3, where this capture was treated as a two-step process. In the first step, there is an exchange of neutrinos, which occurs by virtue of a weak interaction which does not conserve lepton charge and which leads to a mixing of the initial and excited daughter atomic states. In the second step, the atom decays through the emission of x rays and/or electrons, while the nucleons decays through the emission of an internal-bremsstrahlung $\gamma$ ray, which carries off either all the excitation energy or the difference between this excitation energy and the energy of the excited level of the product nucleus. The detection of these $\gamma$ rays is a characteristic feature of specifically a neutrinoless process, which $\gamma$ transitions between levels of the daughter nucleus could be associated with not only $0\nu$ capture channels but also $2\nu$ capture channels. Calculations of $T_{1/2}$ have been carried out only for $^{58}$Ni, $^{92}$Mo, $^{96}$Ru, (Ref. 2), and $^{112}$Sn (Ref. 3). They predict values of $10^{24}$–$10^{25}$ yr for the $2\nu$ decay channel[2,3] and $10^{30}$–$10^{36}$ yr (Ref. 2) or $10^{22}$–$10^{26}$ yr (Ref. 3) for the $0\nu$ channel.

One object of interest in the search for the $2e$ capture is the nucleus $^{196}$Hg, which converts into $^{196}$Pt in the course of the capture (Fig. 1). The abundance of $^{196}$Hg is[7] 0.15%, and the difference between the masses of the $^{196}$Hg and $^{196}$Pt atoms is[8] $820 \pm 3$ keV. The excitation energy available in the $0\nu 2K$ capture of $^{196}$Hg ($663.2 \pm 3$ keV) can be carried off by a single $\gamma$ ray or by two $\gamma$ rays, with energies of 307.5 and 355.7 keV.

The present experiment was carried out with a low-background semiconductor spectrometer[9] and involved measurement of the background of a Ge(Li) detector surrounded by a mercury shield with a mass of 320 kg (480 g of $^{196}$Hg). The apparatus was in an underground laboratory in a salt mine 430 m below ground level.[10] The Ge(Li) detector has a volume of 35 cm$^3$ and a resolution of 3.8 keV at the 1332-keV line. The cryostat of the detector is mode of "especially pure" titanium and copper.

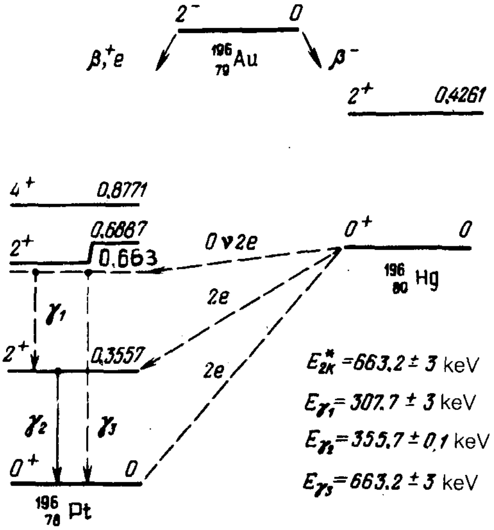

FIG. 1. Level scheme of the triplet $^{196}$Hg–$^{196}$Au–$^{196}$Pt.

The detector is shielded from the background by layers of mercury (8 cm thick), lead (10 cm), and polyethylene (16 cm). The goal is to reduce the detector background to an extremely low level. The intensity of the $\gamma$-ray peak of $^{208}$Tl (2615 keV), for example, is $(8 \pm 4) \times 10^{-3}$ count/h, and the average count rate in the interval 2020—2060 keV is $(1.7 \pm 0.5) \times 10^{-4}$ count/h · keV) (Ref. 11).

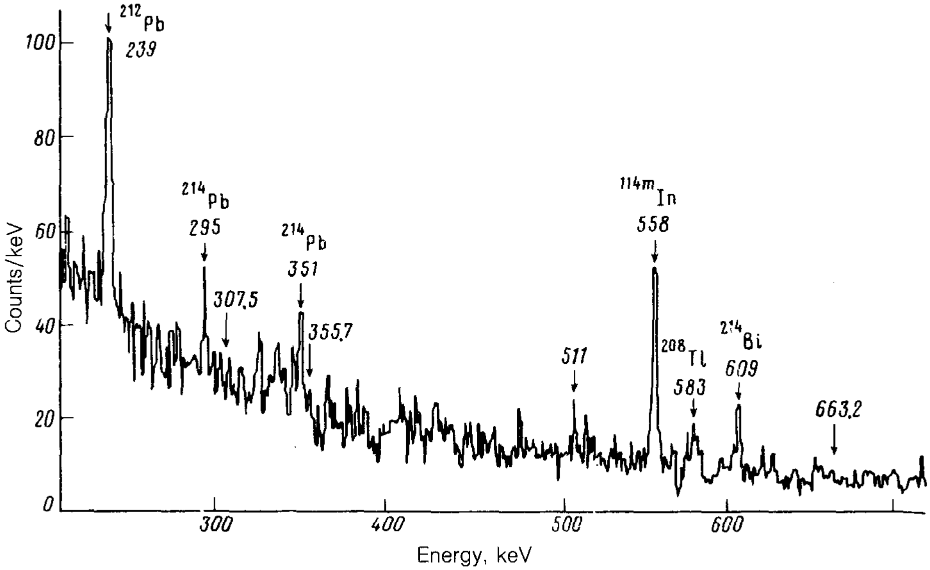

FIG. 2. Background spectrum of the Ge(Li) detector measured in an underground experiment lasting 1478 h.

The experiment was carried out from December 1984 to July 1985. The measurement procedure and the procedure used to process the data are described in detail in Refs. 9 and 11.

The region studied in the overall background spectrum of the detector, recorded over 1478 h, is shown in Fig. 2 at an energy scale of 1 keV/channel. The $\gamma$-ray lines in this spectrum belong for the most part to the daughter ratio nuclides of the $^{232}$Th and $^{238}$U natural radioactive series. The average background levels in the regions in which we would expect to find peaks from $2K$ capture in $^{196}$Hg are $(1.9 \pm 0.1) \times 10^{-2}$, $(1.4 \approx 0.1) \times 10^{-2}$ and $(4.9 \pm 0.3) \times 10^{-3}$ count/(h · keV) at energies of 310, 360, and 660 keV, respectively.

The absence of an effect means that we can set limits on the branching ratio for $2e$ capture in $^{196}$Hg on the basis of this experiment. To find these limits, we need to know the limiting value, for the given background level, of the intensities of the $\gamma$ peaks being sought and the absolute efficiency at which the $\gamma$ rays emitted by $^{196}$Hg atoms are detected. These atoms are distributed uniformly through the mercury shield around the detector. The efficiency was calculated by the program of Ref. 12, which takes into account the measurement geometry, the absorption of $\gamma$ radiation, and the intrinsic detection efficiency of the detector. The calculation procedure was tested in measurements with point and bulk $^{137}$Cs and $^{40}$K sources. The discrepancy between the calculations and experiment did not exceed 10%. The calculated values of the absolute efficiency for the detection of $\gamma$ rays from the 2 K capture of $^{196}$Hg increase from $6.5 \times 10^{-6}$ (300 keV) to $1.2 \times 10^{-5}$ (600 keV).

To evaluate the limiting intensity of the $\gamma$-ray peaks of the 2 K capture of $^{196}$Hg

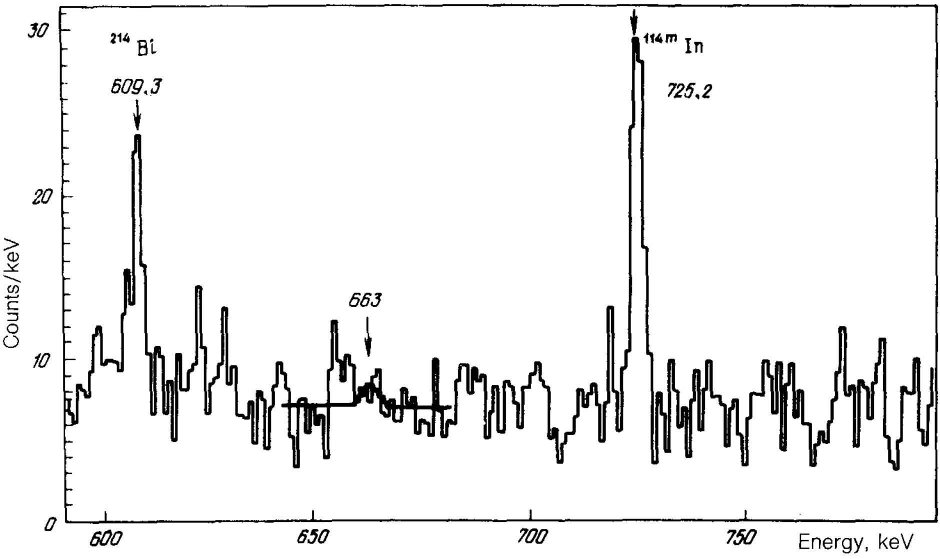

FIG. 3. Part of the background spectrum and the approximating function for the interval 643–683 keV.

TABLE I. Experimental limits on $T_{1/2}$ with respect to $2e$ capture in the nuclei $^{58}$Ni, $^{64}$Zn, $^{92}$Mo, $^{106}$Cd, and $^{196}$Hg.

| Transition, $M(A, Z) - M(A, Z-2)$, keV | Study | Detector | Energy of $\gamma$ ray, keV | Limit on $T_{1/2}$, yr | |
|---|---|---|---|---|---|
| | | | | $0\nu 2K$ capture | $(0\nu + 2\nu)2e$ capture |
| $^{58}$Ni – $^{58}$Fe<br>1927,2 ± 0,7 | [6] | 2NaI(Tl)<br>Ø 15 · 10 см$^2$ | 1927 | 2,1 · $10^{19}$ | — |
| | [5] | Ge(Li) 125 см$^3$ | 810.8 (2$^+$)<br>1674,7 (2$^+$) | — | 4 · $10^{19}$<br>4 · $10^{19}$ |
| $^{64}$Zn – $^{64}$Ni<br>1096,3 ± 0,9 | [4] | Proportional counter | 7.48 (x radiation) | — | 8 · $10^{17}$ |
| $^{92}$Mo – $^{92}$Zr<br>1648 ± 4 | [5] | Ge(Li) 125 см$^3$ | 934.5 (2$^+$)<br>1383 (0$^+$)<br>1495,6 (4$^+$) | — | 3 · $10^{18}$<br>4 · $10^{18}$<br>6 · $10^{18}$ |
| $^{106}$Cd – $^{106}$Pd<br>2778 ± 8 | [6] | 2NaI(Tl)<br>Ø 15 · 10 см$^2$ | 2781 | 1.5 · $10^{17}$ | — |
| $^{196}$Hg – $^{196}$Pt<br>820 ± 3 | Present study | Ge(Li) 35 см$^3$ | 307.5<br>355.7 (2$^+$)<br>663,2 | 1,3 · $10^{17}$<br>2.5 · $10^{17}$ | 1.5 · $10^{17}$ |

which are possibly present in the background spectrum, we approximated the experimental data near the expected $\gamma$ lines ($\pm 20$ keV) by the method of least squares as the sum of two functions, representing the background (a linear function) and the effect (a Gaussian function with a given half-width and a given position of the center of gravity). Figure 3 shows part of the background spectrum, along with the approximating function for the interval 643–683 keV. The values found for the area under the Gaussian curve in the course of the fit and the error in this area were used to estimate $T_{1/2}$ for $^{196}$Hg with a given confidence level. For this estimate we took into account the measurement time, the number of $^{196}$Hg nuclei ($1.4 \times 10^{24}$), and the detection efficiency. This fitting procedure was repeated after the center of gravity of the Gaussian curve was moved in 0.5-keV steps over an interval of $\pm 4$ keV. Since the resulting estimates of $T_{1/2}$ varied insignificantly ($\pm 15\%$) we adopted as the final values the average values of the limits on $T_{1/2}$, which are as follows, at a confidence level of 68%:

$$T_{1/2}\begin{cases} 1.5\times 10^{17}\ \text{yr for } 2e \text{ capture to the 355.7-keV level,} \\ 1.3\times 10^{17}\ \text{yr for } 0\nu 2K \text{ capture } (E_\gamma = 307.5\ \text{keV}), \\ 2.5\times 10^{17}\ \text{yr for } 0\nu 2K \text{ capture } (E_\gamma = 663.2\ \text{keV}). \end{cases}$$

The experimental data found previously[4-6] (Table I) for the nuclei $^{58}$Ni, $^{64}$Zn, $^{92}$Mo, and $^{106}$Cd and also the results of the present experiment show that the sensitivity level achieved in these experiments ($T_{1/2} \approx 10^{17}$–$10^{19}$ yr) is still far from the theoretical estimates ($T_{1/2} \approx 10^{22} - 10^{36}$ yr). We note, however, that the theoretical estimates are ambiguous; calculations have not been carried out for $^{196}$Hg, $^{106}$Cd, or $^{64}$Zn; and the procedures used in the experiments which have been carried out so far have been oriented toward detecting single $\gamma$ rays, while the total signal from the $0\nu 2e$ capture would have unique properties, consisting of the simultaneous emission of two x rays and a $\gamma$ ray with an energy different from the energies of $\gamma$ transitions of the daughter nucleus. The development of a highly efficient apparatus for measuring all the characteristics of such a signal could, even in the foreseeable future, raise the sensitivity of the experimental search for $0\nu 2e$ capture to the level of the theoretical predictions. The latter also require refinements.

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