Search for Double Beta-Decay in Sn^{124} and Zr^{96}†,*

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If double beta-decay takes place without the emission of neutrinos, the total kinetic energy of the two emitted electrons will be constant. The half-life would be of the order of 10^{16} years for 1.5-Mev available energy or 10^{14} years for 3.5-Mev available energy.

Sn^{124} and Zr^{96} have been studied for evidence for such a process. A lower limit of 1.5×10^{17} years is set on the half-life for double beta-decay of Sn^{124}, which has 1.5 ± 0.4-Mev available energy. In the case of Zr^{96}, which has 3.4 ± 0.3-Mev available energy, the results indicate a lower limit of 2×10^{16} years for double beta-decay. The results also indicate the presence of a slight activity of $6\pm2\times10^{16}$ years half-life (if attributed to Zr^{96}) at 3.8 ± 0.5 Mev with an energy spectrum and coincidence nature compatible with the predictions for double beta-decay. Further experimentation is necessary to establish the existence and nature of this activity.

INTRODUCTION

THE existing theories dealing with double beta-decay fall into two general classes. One theory[1] requires the emission of two neutrinos and two electrons and predicts a lifetime of the order of 10^{22} years for 4-Mev available energy. The second class of theories[2–4] allows the emission of two electrons unaccompanied by neutrinos and predicts lifetimes of the order of 10^{14} years for 3.5-Mev available energy. If the latter theory is correct, the total kinetic energy of the two electrons will be constant, and it might be possible, because of this property, to detect double beta-decay directly.

The most recent theoretical treatment of double beta-decay without the emission of neutrinos is that of Primakoff.[3] Using the tensor interaction and shell model predictions for even-even nuclei (spin 0, even parity) he finds an angular correlation between the electrons of approximately $(1+\cos\theta)$ and predicts lifetimes of 6×10^{15} years for 1.5-Mev available energy and 6×10^{13} years for 3.5-Mev available energy.

These predicted lifetimes are based on constants whose values are calculated from experimental data on the decay of the neutron. Experiments on the beta-decay of heavier nuclei indicate that the lifetimes may be much longer, since the transitions are probably unfavored.[5,6]

THE EXPERIMENT

If double beta-decay occurs without the emission of neutrinos, the total kinetic energy of the two emitted electrons will be constant, as noted above. The experiment reported here takes advantage of this fact. The sample to be investigated was placed between two *trans*-stilbene crystals observed by separate photomultipliers. Simultaneous current pulses from these photomultipliers were added electronically and analyzed by a thirty-channel pulse-height discriminator.[7] During the coincidence runs, recording occurred only when there was a coincidence between pulses from the two photomultipliers. In view of the possibility of a strong angular correlation between the emitted electrons, runs were also made measuring total activity without regard to coincidences. An anticoincidence system of four large guard counters was used to reduce the background. The equipment was shielded by $\frac{1}{4}$ inch of steel inside 4 inches of lead. Thermal neutrons were absorbed by a $\frac{1}{16}$-inch cadmium sheet under the shielding and 1 inch of borax around it.

Trans-stilbene scintillation crystals observed by RCA 5819 photomultiplier tubes were used throughout this experiment. The experimental arrangement is

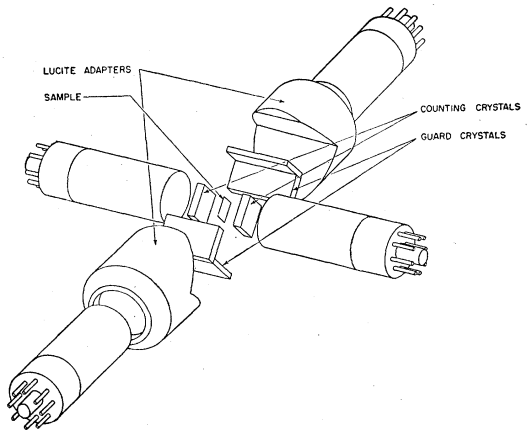

FIG. 1. Expanded schematic view of the counting and anticoincidence unit.

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[1] M. Goeppert-Mayer, Phys. Rev. **48**, 512 (1943).
[2] W. H. Furry, Phys. Rev. **56**, 1148 (1939).
[3] H. Primakoff, Phys. Rev. **85**, 888 (1952).
[4] L. A. Sliv, Zhur. Eksptl. i Teort. Fiz. **20**, 11, 1029 (1950).
[5] H. Primakoff, private communication, July 8, 1952.
[6] J. A. McCarthy, thesis, University of Rochester, Rochester, New York, 1952 (unpublished).
[7] Fulbright, McCarthy, and McCutcheon, Phys. Rev. **87** 184 (1952).

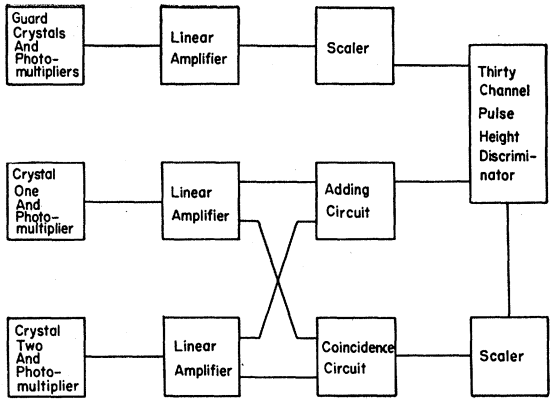

FIG. 2. Block diagram of electronic equipment.

shown in Fig. 1 and the electronic equipment in Fig. 2. The sample under investigation was placed between two crystals (each 2.5×2.5×1.1 cm, placed 3 mm apart) which were observed by separate photomultipliers whose outputs were added electronically. These shall be called "counting crystals" in the future to avoid confusion. The source and the counting crystals were surrounded on four sides by guard crystals whose outputs were in anticoincidence with the added pulse from the counting crystals. The same crystals were used to investigate all samples. To permit sample changing one counting crystal and photomultiplier were made removable.

The sample under investigation and the appropriate background sample were alternately placed between the counting crystals for periods of from ten to twenty hours. Such rotation of samples was used throughout the entire experiment. During the necessary handling of the samples, clean silk gloves were worn to prevent possible contamination.

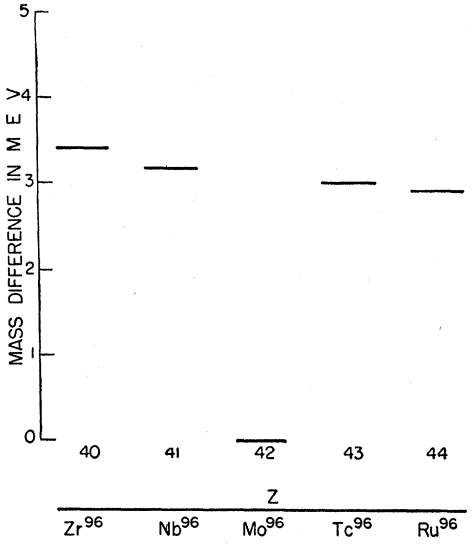

FIG. 3. Atomic mass differences above most stable nuclide of isobaric series $A=96$.

Separate energy calibrations of the two counting crystals were made approximately every 48 hours, using the gamma-rays from Na^{22}. The operation of the thirty channel pulse-height discriminator was checked every twelve hours. If any change in gain or any electronic difficulty was found in one of these inspections, all results since the last satisfactory check were discarded.

The isotopes investigated were Sn^{124} and Zr^{96}.[8] Figures 3 and 4 show the comparative atomic masses of the members of the isobaric series $A=96$ and $A=124$, from beta-decay data[9–11] and from recent mass-spectrographic measurements.[12,13]

The masses given represent probable values, and are in general accurate only to about 0.5 Mev.

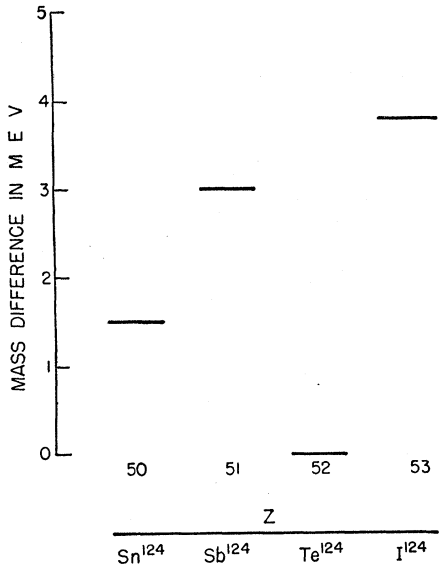

FIG. 4. Atomic mass differences above most stable nuclide of isobaric series $A=124$.

EXPERIMENTAL RESULTS

The experimental results are shown on Figs. 5 to 9. Sample statistics bars are given to show the order of magnitude of the probable error resulting from the random counting rate. Each statistics bar is the square root of the number of counts in the interval. The activity measured with Zr^{96} or Sn^{124} is shown on the graphs as a solid line, while the activity measured with the comparison samples is shown with a dotted line.

[8] The enriched samples used were supplied by the Oak Ridge National Laboratory. The Sn^{124} metal sample weighed 150 milligrams and contained 95 percent Sn^{124}; the Zr^{96} sample weighed 52 milligrams and contained 89.5 percent Zr^{96}; the Zr^{94} sample weighed 52 milligrams and contained 97.9 percent Zr^{94} (the Zr samples were in the form of ZrO_2, a powder).

[9] *Nuclear Data*, National Bureau of Standards Circular 499 (1950).

[10] P. Preiswerk and P. Stähelin, Helv. Phys. Acta **24**, 300 (1951).

[11] G. E. Boyd and B. H. Ketelle, Oak Ridge National Laboratory Reports ORNL 795 (1951) and ORNL 879 (1951).

[12] B. G. Hogg and H. E. Duckworth, Phys. Rev. **86**, 567 (1952).

[13] H. E. Duckworth, private communication, May 26, 1952.

If double beta-decay occurs without the emission of neutrinos, it should be observed as a peak in the Zr^{96} or Sn^{124} activity above the activity measured with the corresponding background sample in both total activity and coincidence data. In Sn^{124} this peak should occur at 1.5 ± 0.4 Mev;[12] in Zr^{96} at 3.4 ± 0.3 Mev.[13]

A. Sn^{124}

Figure 5 shows the coincidence activity in Sn^{124} and in natural Sn. No statistically significant difference between the two samples is observed. In the region from one to two Mev, the excess of Sn^{124} over natural Sn is -15 ± 26 counts in 110 hours.

Figure 6 shows the results of 128.3 hours each of Sn^{124} and natural Sn samples with the equipment set to record total activity without regard to coincidences. A small excess activity in the Sn^{124} sample between 1.9 and 3.5 Mev is indicated. Below 1.9 Mev the background is so high that any small excess would not be statistically significant. The excess activity between 1.9 and 3.5 Mev is 2.6 ± 0.6 counts per hour.

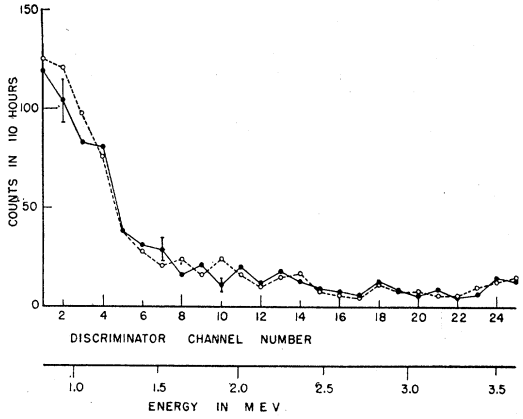

FIG. 5. Coincidence activity with Sn^{124} and natural Sn samples. The solid line is Sn^{124} and the dashed line is natural Sn.

B. Zr^{96}

The results of a similar total activity measurement on Zr^{96} are shown in Fig. 7. In the region between 1.3 and 2.2 Mev, which is not shown, the Zr^{96} sample causes an excess of 0.45 ± 0.34 counts per hour. The apparent peak between 3.3 and 4.3 Mev indicates an activity in Zr^{96} with a counting rate of 0.67 ± 0.18 counts per hour.

Figure 8 gives the results of a coincidence activity experiment designed to study the nature of the 3.8-Mev peak in Zr^{96} indicated by the total activity portion of this experiment. Between 3.3 and 4.3 Mev, Zr^{96} shows an excess of 0.25 ± 0.09 counts per hour.

Figure 9 shows the difference between the Zr^{96} results and the corresponding Zr^{94} background results for both total and coincidence activity. It is included to illustrate that the evidence for a peak at 3.8 Mev is not as conclusive as might be assumed from an inspection of Fig. 7.

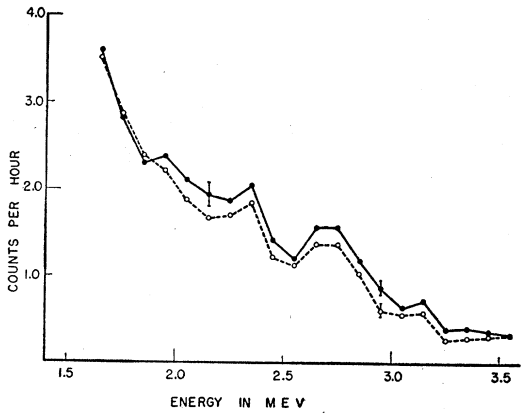

FIG. 6. Total activity with Sn^{124} and natural Sn samples. The solid line is Sn^{124} and the dashed line is natural Sn.

DISCUSSION

A. Sn^{124}

In the case of Sn^{124} no evidence is found for double beta-decay. No evidence for the "neutrino-less" theory can be found in this or any other experiment[14–17] which has been performed on Sn^{124}. However, we set a lower limit of 1.5×10^{17} years on the lifetime of Sn^{124} for double beta-decay (assuming no angular correlation between the emitted electrons).

In the experiment on Sn^{124} which measured total counting rate, an activity extending to more than 3 Mev is indicated. This cannot be accounted for by simple beta-decay of Sn^{124} to Sb^{124} and thence to Te^{124}. Levine and Seaborg[18] searched for evidence of such a

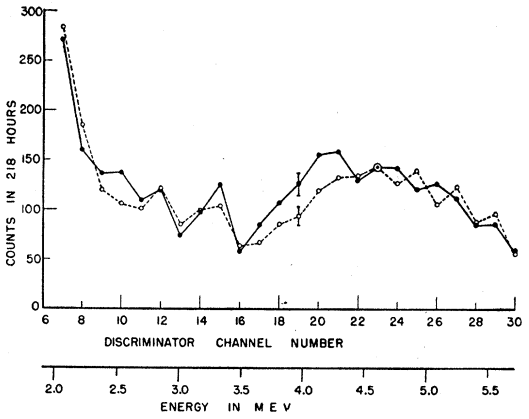

FIG. 7. Total activity with Zr^{96} and Zr^{94} samples. The solid line is Zr^{96} and the dashed line is Zr^{94}.

[14] J. S. Lawson, Phys. Rev. **81**, 299 (1950).
[15] M. I. Kalkstein and W. F. Libby, Phys. Rev. **85**, 368 (1952).
[16] E. L. Fireman and D. Schwarzer, Phys. Rev. **86**, 451 (1952).
[17] R. M. Pearce and E. K. Darby, Phys. Rev. **86**, 1049 (1952).
[18] C. A. Levine and G. T. Seaborg, University of California Radiation Laboratory Report UCRL 635.

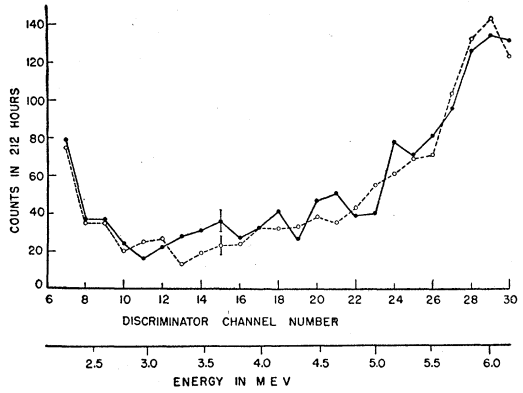

FIG. 8. Coincidence activity with Zr^{96} and Zr^{94} samples. The solid line is Zr^{96} and the dashed line is Zr^{94}.

process and set a lower limit on the lifetime of 3.7×10^{17} years. This is in agreement, of course, with mass spectrographic and beta-decay data which indicate that such a decay is forbidden by conservation of energy. It is extremely difficult to explain the observed activity unless the sample is contaminated.

B. Zr^{96}

Since Zr^{96} has 3.4-Mev available energy, it is a favorable isotope for study, with a predicted lifetime of about 6×10^{13} years.

In the 218-hour run measuring total activity without regard to coincidences (Fig. 7), 816 ± 29 counts were recorded between 3.3 and 4.3 Mev with the Zr^{96} sample and 669 ± 26 counts with the Zr^{94} sample, indicating an activity which, if arising from Zr^{96}, has a counting rate of 0.67 ± 0.18 counts per hour. Slight excess in the Zn^{96} sample was also noted at energies below 2.2 Mev with no excess from 2.3 to 3.3 Mev or above 4.3 Mev. Since there is apparently 0.3 ± 0.3 Mev available for direct beta-decay of Zr^{96} to Nb^{96}, this low energy excess might be explained by simple beta-decay with a lifetime of the order of 10^{15} years. The peak at 3.8 Mev cannot be accounted for by beta- or gamma-activity because there is no evidence of activity between 2.3 and 3.3 Mev. The possibility of an internally converted 3.8-Mev gamma-ray exists, but there is no evidence of Compton electrons in the lower energy region. In order to account for an internal conversion peak coupled with a number of emitted gamma-rays small enough to be compatible with the apparent absence of Compton electrons originating in the crystal, it would be necessary to assume that the 3.8-Mev gamma-ray was at least 5 percent internally converted, which is a factor of more than 10^3 higher than the predicted internal conversion coefficient.[19] The sample with its aluminum foil covering is 35 milligrams/cm² thick, so any alpha-particle activity would also be smeared over the entire low energy region.

[19] S. Dancoff and P. Morrison, Phys. Rev. **55**, 133 (1939).

Thus, from this part of the experiment we have four possibilities for the 3.8-Mev peak:

1. A statistical effect. The probability of such an effect is less than 1 percent.
2. Internal conversion of a 3.8-Mev gamma-ray with an extraordinarily large internal conversion coefficient.
3. Internal pair production.
4. Double beta-decay with a lifetime of $0.6\pm0.2\times10^{17}$ years.

In view of the coincidence data presented below, the author considers the fourth hypothesis (double beta-decay) the most likely one.

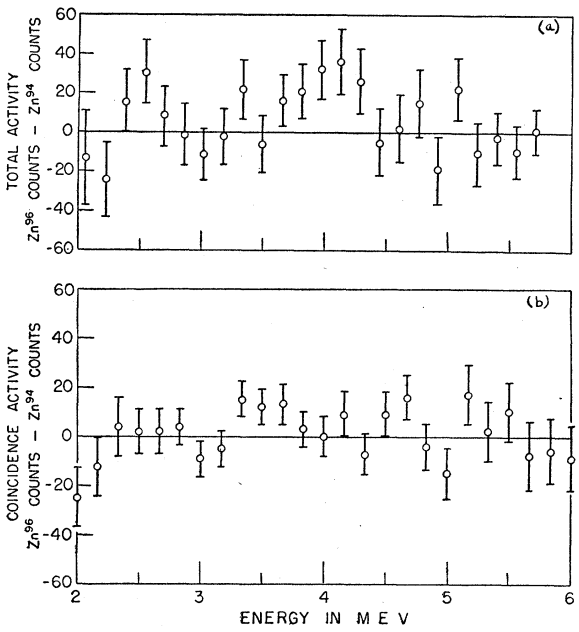

FIG. 9. Difference between activity with Zr^{96} sample and with Zr^{94} sample. The difference in the total activity in 218 hours is shown in the upper diagram; the difference in the coincidence activity in 212 hours is shown in the lower diagram. The energy scale is the same for both. Each error is the square root of the sum of the Zr^{96} and the Zr^{94} counts in the interval.

The peak of about 5 counts per hour with both Zr^{96} and Zr^{94} samples between 4 and 5 Mev is probably high energy cosmic-ray particles crossing both counting crystals. In the coincidence data presented below one of the counting crystals was replaced by a crystal 8 millimeters thicker and the peak moved to a position above 5 Mev.

The Zr^{96} experiment was repeated, using coincidences between the counting crystals. The results, shown in Fig. 9, show an excess with the Zr^{96} sample of 0.25 ± 0.09 counts per hour between 3.3 and 4.3 Mev. Unfortunately, the geometry of the experiment, the large statistical errors, and the probability of single or multiple scattering in the sample and crystals prevent any conclusion regarding the angular correlation of the emitted electrons.

CONCLUSION

In view of the large statistical errors, the author feels that the results presented here indicate, but do not prove, that double beta-decay may occur in Zr^{96} without the emission of neutrinos. Further experimentation is clearly necessary.

The author wishes to thank Professor H. W. Fulbright for suggesting this experiment and for his advice and encouragement both before and during the performance of the research. He also wishes to thank Mr. A. Petschek for many helpful discussions regarding the theoretical background of the problem.

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Argon 38 in Pitchblende Minerals and Nuclear Processes in Nature

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The isotopic constitution of argon extracted from four pitchblende samples of different age and uranium concentration has been determined. The A^{36}/A^{38} ratio was found to vary by over three hundred percent with the greatest deviations from normal occurring in the ores with high uranium content. It is suggested that A^{38} has been produced in considerable quantities in these ores by nuclear reactions involving α-particles and/or the spontaneous fission neutrons.

THE occurrence of large variations in the abundance of A^{40}, because of K capture in K^{40}, in argon samples extracted from rocks is well known and has been studied in many laboratories for the purpose of age determinations. For example, Mousuf[1] has found values of the A^{40}/A^{36} ratio as high as 90 000 in samples of microcline while the value of this ratio for normal argon, as determined by Nier,[2] is 295.5. Since no detailed study of the A^{38} content of argon samples from various sources has been reported, a survey of the A^{38} abundance in pitchblende ores was undertaken in this laboratory in conjunction with the investigation of the xenon and krypton spontaneous fission yield patterns already in progress.[3]

Powdered pitchblende samples of approximately 200 grams were placed in an inconel combustion tube and continuously evacuated for 24 hours and then heated to 250–300°C for one hour to remove most adsorbed gases. Preliminary experiments showed that no loss of the fission product gases occurred during this period. The temperature was then slowly raised to the maximum obtainable with the furnace used (approximately 1250°C) and the gases released were collected in two or three fractions over the temperature range covered. Fractions collected at the higher temperatures should contain less normal argon from adsorbed gases. Each fraction was purified in a calcium furnace to constant volume. The volume of the individual samples varied from one to ten cubic millimeters depending on the ore used and the temperature range covered.

The samples were analyzed on a 180° direction focusing mass spectrometer. The small size of the samples made it necessary to operate the ion source with very high ionizing electron beam currents.

The principal sources of error in determining the abundance of a rare isotope, such as A^{38}, are impurities in the sample and residual or background ion currents in the mass spectrometer. To eliminate the first factor one sample was removed from the mass spectrometer and repurified in a calcium furnace. The analyses before and after repurification were in good agreement (Table I). Increasing the peak heights by a factor of three by changing the sample pressure produced no detectable change in the abundance ratios, indicating that the effect of the background ion currents was negligible.

The results of the mass spectrometer analysis are given in Table I together with the ratios for normal argon. The uranium concentration and geological ages determined by lead isotope ratio measurements are also listed. The Great Bear Lake pitchblende result is from preliminary work and only the highest temperature fraction was available for analysis. The A^{36}/A^{38} ratio is seen to vary from 1.69 to 5.18 for the pitchblende samples, compared with 5.35 for normal argon. This is an over-all variation of more than 300 percent. Such large changes in isotopic abundance cannot be explained by any natural fractionation process and can be produced only by some nuclear reactions. The xenon and krypton fission yield patterns were found to be independent of the temperature at which the samples were collected, indicating that no appreciable fractionation results from the diffusion of the gases out of the crystal lattice during the extraction process.

It is, therefore, necessary to consider possible nuclear reactions which would result in the production of A^{38} or the removal of A^{36}. Since no correlation between the

[1] A. K. Mousuf, Phys. Rev. **88**, 150 (1952).
[2] A. O. Nier, Phys. Rev. **77**, 789 (1950).
[3] J. Macnamara and H. G. Thode, Phys. Rev. **80**, 471 (1950).