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Search for Double Beta Decay in Cadmium and Molybdenum*

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Random Wilson cloud-chamber photographs of cadmium and molybdenum foils show that the double negatron half-life is greater than or equal to 1×10^{17} years in Cd^{116} and 3×10^{17} years in Mo^{100}, and that the double positron half-life is greater than or equal to 6×10^{16} years in Cd^{106} and 4×10^{18} years in Mo^{92}. The background appears to be primarily caused by Compton electrons and photoelectrons ejecting other electrons on their way out of the foil and by photons that suffer two Compton scatterings while traversing the foil.

I. INTRODUCTION

Several paths to an experimental choice between the Dirac and Majorana neutrino theories are, in principle, possible. For instance, discovery of a neutrino magnetic moment[1] would disprove the Majorana model because electromagnetic forces would take the neutrino wave function out of the Majorana subspace.[2] On the other hand, if neutrinos from the sun contributed to the background in the inverse beta experiments of Reines and Cowan,[3] or if pile neutrinos could cause negatron emission from stable nuclei,[4] then neutrinos and antineutrinos would be equivalent in beta decay. This equivalence would probably imply the correctness of the Majorana theory, although there are alternative formalisms.[5,6] Also, the shape of the electron energy spectrum in mu-meson decay might depend on whether the two neutrinos emitted are identical or not. However, the spectrum observed seems to be consistent with either identical or different neutrinos.[7]

The most promising procedure for distinguishing between the neutrino theories is still an investigation of double beta-decay. If neutrinos and antineutrinos are not equivalent, double beta decay would involve the emission of two electrons and two neutrinos or antineutrinos, and half-lives in excess of 10^{21} years would result.[8] If neutrinos and antineutrinos are equivalent, there needs to be only one virtual neutrino which has all phase space available. Then one would expect half-lives ranging upwards from 10^{13} years depending on the form of the interaction, the energy, and the nuclear matrix elements.[9,10] If the decay is primarily to a single final state, as it should be because of the strong energy dependence of the transition probability, the sum of the energies of the electrons emitted will be constant. Unless one admits fairly artificial theories,[5,6,11,12] a demonstration of such a constant sum of energies would confirm the Majorana theory.

Lower limits ranging from 10^{14} years through 10^{19} years have been set for double beta activities of more than twenty nuclides in several investigations.[12–19] Such lower limits can never refute the Majorana theory because a large decrease in transition probabilities can result if the overlap of final and initial states is poor. The accumulation of such lower limits, however, particularly if they are above 10^{16} years, does decrease the likelihood of the neutrino being a Majorana particle.

There exists very little positive evidence for double beta decay. The Sn^{124} results of Fireman[20] have been refuted by various investigators, including Fireman himself.[19] Inghram and Reynolds[21] found evidence for the decay of Te^{130} with a half-life of about 10^{21} years, but, as has already been pointed out by Kohman,[18] the activity discovered could well be single beta decay. Fremlin and Walters[16] found a 10^{15}-year activity in Mo^{100}, but Kohman[18] found a lower limit of 10^{16} years and the present work gives a lower limit of 3×10^{17} years. McCarthy[22,23] found activities in Zr^{96} and Ca^{48} and showed that, although there are alternatives, they most probably represent double beta decays in which the two emitted negatrons carry away all of the available energy with a half-life of 6×10^{16} years. Clearly, more conclusive results are prerequisite to a final decision between Dirac and Majorana neutrinos.

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II. SELECTION OF SOURCES

Among the elements which might show double beta activity, cadmium and molybdenum seem particularly suitable for experimental investigation. The nuclide Cd^{116}, with natural abundance of 7.7 percent, has 2.6 ± 0.2 Mev, and Mo^{100}, with a natural abundance of 9.7 percent, has 2.3 ± 0.2 Mev available for double negatron emission.[24,25] In addition, Cd^{106}, with a natural abundance of 1.2 percent, has 0.8 ± 0.2 Mev available for the kinetic energy of the positrons in double positron emission.[24,25] Molybdenum also contains, with a natural abundance of 15 percent, Mo^{92}, which lies below the beta stability line and might undergo double positron decay; however, since it has a magic number of neutrons and since no adequate mass measurements exist, nothing reliable can be said about the energy available. If the neutrino is a Dirac particle, the half-lives for all four transitions are too long to be observed in the procedure described below. If the Majorana theory applies, the half-lives for Cd^{116} and Mo^{100} will be $\geq 10^{15}$ years, and the half-life of Cd^{106} will be $\geq 10^{16}$ years. Since the energy is not known for Mo^{92}, a lifetime cannot be estimated; in fact, only double K capture, or even no transition at all, may be energetically possible.

III. EXPERIMENTAL PROCEDURE

For both the cadmium and the molybdenum investigations, three foils of the material being studied were stretched across a 24-cm diameter Wilson cloud chamber. A mixture of 50 percent by volume of water and 50 percent methanol was boiled into the evacuated chamber, which was then filled to a total pressure of 1.6 atmos with argon. Stereoscopic photographs were taken of a random expansion every 30 seconds. A magnetic field of 790 gauss, made uniform to 1.5 percent over the usable region of the chamber by proper shaping of an air-core solenoid,[26] was turned on about one second before the expansion. The adjustment of the chamber was checked visually about every hundred expansions.

The pictures obtained were projected first on a screen, with the two stereoscopic views placed next to each other. Those in which two negatrons or two positrons seemed to come from the same point on the foil were marked for further study. Also, from the number and appearance of single tracks, a judgment of the acceptability of each roll of film was made: 13 percent of the photographs obtained were discarded because the appearance of the tracks and the background fog suggested doubt whether minimum-ionization electron tracks could be discerned in all parts of the chamber.

The pictures marked for further examination were projected stereoscopically through the same mirror and camera system with which they were photographed. By attempting to place a screen in the plane defined by the beginnings of both tracks, one could determine with fair precision whether the tracks came from the same point. Some difficulty arose because the layer of gas adjacent to the foil loses its supersaturation sooner than most of the chamber, so that many tracks became invisible in that layer and had to be extrapolated back to the foil. In all cases, however, it could be determined whether the tracks started within 0.4 cm of each other. When both tracks appeared to come from the same point, upper and lower bounds on their curvature, and therefore on their energy, were established. When one of the tracks was scattered too violently in the gas or was too nearly straight for meaningful measurement, the picture was discarded. Therefore, only tracks of electrons with energy between 0.1 Mev and 6 Mev could be used. For those photographs in which both tracks came from the same point and had a measurable curvature, an upper and lower bound on the sum of the energies of the two electrons, including a correction for loss in the foil, was established.

IV. CADMIUM RESULTS

For the cadmium investigation, 30 g of 0.11 g/cm² foil,[27] containing 1.2×10^{22} atoms of Cd^{116} and 1.9×10^{21} of Cd^{106}, were installed in the chamber. The probability that two electrons from one atom would travel in the illuminated region of the chamber for 4 cm was, on the average, 0.3, giving 3.6×10^{21} as the effective number of Cd^{116} nuclei and 5.7×10^{20} as the effective number of Cd^{106} nuclei.

With the foils in place and with the chamber filled and operated as for the actual double beta search, a calibrated P^{32} source was photographed. From the strength of the source and a determination of the average number of tracks seen coming from it per expansion, the sensitive time of the chamber was found to be 0.36 ± 0.08 second per photograph. Since 12 352 acceptable photographs were taken, the cadmium was examined for $(1.4\pm0.3)\times10^{-4}$ year.

In 24 cases, two negatron tracks appear to originate from the same point in the foil. To represent the results in Fig. 1, a rectangle of unit area was assigned to each event, with its base running from the minimum to the maximum total energy of the event. The solid line in Fig. 1 was obtained by adding the heights of these rectangles wherever they overlapped and gives the experimental number per unit energy as a function of the energy.

A definite indication of double beta decay would be a maximum in Fig. 1 at 2.6 Mev. No such maximum is observed, but three events contribute to the result at that location. It is interesting that the calculated background curves (see Sec. VI below) lie everywhere higher then the experimental distributions except at the high-energy end of the cadmium plot. Nevertheless, it is

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clear that the data do not permit a claim of a positive result. Multiplying the effective number of Cd^{116} nuclei by the time for which the cadmium was under examination and by ln2 over three, the number of events that contribute at 2.6 Mev, one finds that the double negatron half-life of Cd^{116} must be $\geq 1\times 10^{17}$ years.

One double positron event was found. Its rectangle, computed as for the negatron events in the aforementioned, is shown dashed in Fig. 1. Its rather large energy range includes the available double positron energy. It is, however, entirely reasonable that the background should contribute one such event (see Sec. VI below). The double positron half-life of Cd^{106} is therefore $\geq 6\times 10^{16}$ years.

V. MOLYBDENUM RESULTS

For the molybdenum investigation, 42 g of 0.13 g/cm^2 foil,[27] containing 2.5×10^{22} atoms of Mo^{100} and 3.9×10^{22} of Mo^{92}, were installed in the chamber. The probability of observing both electrons was again 0.3, giving 7.5×10^{21} and 1.2×10^{22} as the effective numbers of Mo^{100} and Mo^{92} nuclei.

The sensitive time of the chamber was measured with a calibrated P^{32} source as in the cadmium work and was found to be 0.54 ± 0.12 second per photograph. The higher value resulted from the use of a deeper chamber. Since 13 402 acceptable photographs were taken, the molybdenum was examined for $(2.3\pm 0.5)\times 10^{-4}$ year.

In 35 cases, two negatron tracks appear to originate from the same point in the foil. In Fig. 2, the results are shown in the same fashion as for cadmium in Fig. 1. No maximum is observed at 2.3 Mev, the available double negatron energy, but four of the events found straddle that energy.

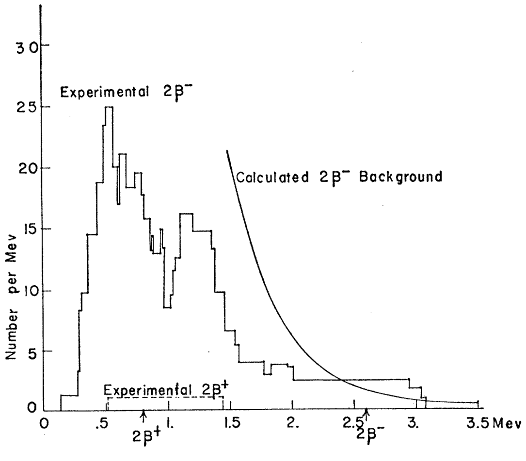

FIG. 1. Number per Mev *vs* energy, of instances in which two electrons appear to come from the same point on the Cd foil. The solid lines give the double negatron distribution. The dotted rectangle represents the one double positron event found. The smooth curve gives the estimated double negatron background. The arrows marked $2\beta^+$ and $2\beta^-$ underneath the energy scale indicate the expected double positron and double negatron energies.

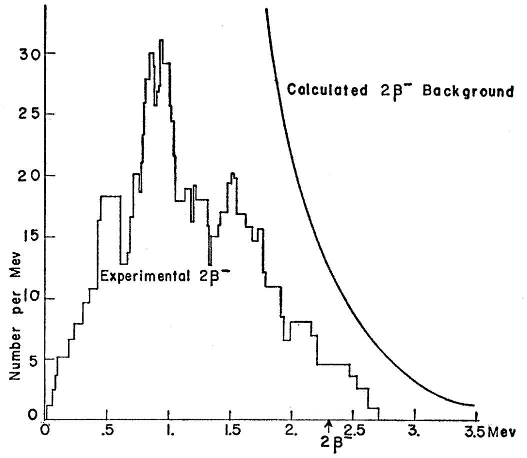

FIG. 2. Number per Mev *vs* energy, of instances in which two negatrons appear to come from the Mo foil. The smooth curve gives the estimated double negatron background. The arrow marked $2\beta^-$ underneath the energy scale indicates the expected double negatron energy.

The calculated background curve (see Sec. VI below) lies everywhere above the experimental results.

Multiplying the effective number of Mo^{100} atoms by the time for which the molybdenum was under examination and by ln2 over four, the number of events that contribute at 2.3 Mev, one finds that the double negatron half-life of $Mo^{100}\geq 3\times 10^{17}$ years.

No double positron events were found. Multiplying the effective number of Mo^{92} atoms by 2.3×10^{-4} year and by 2 ln2, one finds that the double positron half-life of $Mo^{92}\geq 4\times 10^{18}$ year. This result is of little significance in the absence of an estimate of the available energy.

VI. BACKGROUND

Both for the understanding of this experiment and for the planning and interpretation of other double beta experiments, it is useful to study in some detail the sources of background.

For 1869 of the cadmium pictures and 1950 of the molybdenum pictures, all single negatrons that came from the foil (presumably mostly ejected by photons arising from cosmic rays, with perhaps some due to beta-active contamination) were recorded, with their radii of curvature estimated to the nearest cm. From these single negatron distributions were obtained the expected distribution of events in which two electrons appear to emanate from the same point. Figure 3 shows the separate contributions to this background as estimated below for cadmium; for molybdenum, the curves are qualitatively similar. The sums of these contributions are superposed on the experimental distributions in Figs. 1 and 2.

A. Random Coincidence

If the origins of two single tracks happen to be less than 0.4 cm apart, the event can give the appearance

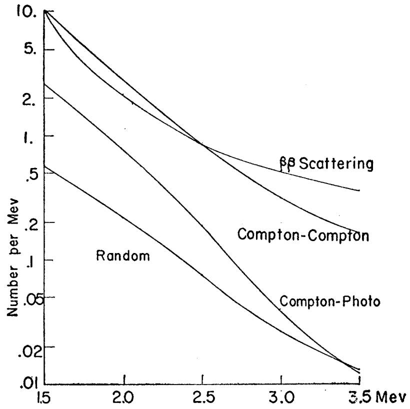

FIG. 3. Contributions in number per Mev to the estimated double-negatron background in Cd, labeled as indicated in Sec. VI.

of a double beta decay. To estimate the contribution of this effect, those pictures containing two or more negatron tracks that start anywhere in the foil were used. All possible combination of the tracks in each picture were considered. Each combination had a probability of 0.4 cm×0.4 cm, the region of distinguishability, divided by 250 cm^2, the total area of the foil. The distribution was then divided by two because tracks formed near the beginning of the sensitive time could be distinguished from those formed near the end by their noticeably more diffuse appearance. The result is the curve labeled *Random* in Fig. 3.

B. Negatron-Negatron Scattering

An event having the appearance of a double beta-decay results if a negatron that has been produced in the foil shares its energy with another negatron on its way out of the foil. Since a negatron starting with 0.3 Mev or less probably would have too little energy after leaving the foil for meaningful measurement, the Møller formula[28] was integrated over the energy shared from 0.3 Mev to half the energy E of the negatron considered. The resulting expression, which will then only depend on E, multiplied by the thickness of foil that the average negatron traverses, and multiplied by the number of negatrons per photograph with E, gives the expected contribution at E. The result is labeled *ββ Scattering* in Fig. 3.

C. Two Compton Scatterings, or Compton Scattering Followed by Photoelectric Emission

Whenever a single negatron was produced by Compton scattering, the residual gamma ray was capable of ejecting another negatron before leaving the foil. It was assumed that all negatrons observed were produced by photoelectric emission or by Compton scattering, and the number due to Compton scattering was computed as a function of energy. The probability that the residual photon would produce another negatron before leaving the foil was then obtained. The calculations were simplified by the assumption that, in each Compton event, the negatron received the average fraction[29] of the photon energy. In Fig. 3, the contribution due to two successive Compton scatterings is labeled *Compton-Compton*; that due to Compton scattering followed by photoelectric emission is labeled *Compton-Photo.*

D. Gas Scattering

If the apparent curvature of either one member of a negatron-positron pair, or of one part of a track going through the foil, is reversed by multiple scattering in the gas, an event giving the appearance of double beta-decay results. The magnitude of this effect was estimated by counting the number of pairs and the number of electrons passing through the foil, and then multiplying by the probability that the curvature of either branch is reversed by scattering.[30] This effect is negligible; in the 2-Mev neighborhood, the expected number of these events is 10^{-3} for either of the entire cadmium or molybdenum experiments.

E. Double Positron Background

An event with the appearance of a double positron decay could result either through gas scattering (see Sec. D), or, far more probably, through two negatrons that start elsewhere converging toward the same point on the foil. The one case found in cadmium is readily explained by the latter process.

F. Summary of Background Calculations

The estimated background suffices to explain the results obtained. Negatron-negatron scattering and two successive Compton scatterings provide the principal contributions.

The sums of all contributions superposed in Figs. 1 and 2 lie, on the whole, somewhat high and rise more rapidly as the energy goes down than the experimental distributions. This behavior might be expected, since no careful consideration of multiple scattering and total absorption of the low-energy electrons in the foil was made.

VII. CONCLUSIONS

It is intriguing that the one double positron event, and the region in which the cadmium double negatron distribution rises above the calculated background, should both occur near the expected energy. Nevertheless, the nature of the background estimates and the smallness of the number of events involved forbid any conclusions other than the setting of lower limits ranging from 10^{16} through 10^{18} years on the half-lives of the processes considered. These lifetimes are consistent

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with both the two-neutrino decay and with unfavored no-neutrino decay.

The background studies indicate limits on the double beta lifetimes that can be reached with expansion chambers.

VIII. ACKNOWLEDGMENTS

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Scarcity of Low-Energy Levels of Be^8 Appearing in Two Boron Reactions

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A careful search is made for evidence of possible excited states in Be^8 in the reactions $B^{11}(p,\alpha)Be^8$ and $B^{10}(d,\alpha)Be^8$, by magnetic momentum analysis at a variety of angles and bombarding energies. In spite of observing the region corresponding to 3 to 8 Mev several times independently under different conditions, with several thousand counts per point on points spaced only about 100 kev apart, no indication was found of any of the states in this region reported by others on the basis of poorer statistics, mostly in other reactions. Each alpha-particle spectrum observed consists of a sharp ground-state peak and a broad peak of the alpha particles giving rise to the well-known 3-Mev excited state of Be^8 superposed on a continuous background from the break-up of this state and from three-particle break-up. Peaks observed near the equivalent of 10 and 11 Mev in Be^8 are identified as arising from a target impurity.

I. INTRODUCTION

THE interesting nucleus Be^8 may be expected to be especially simple because it is so light.[1] It contains just enough nucleons to make up two alpha particles. The nucleus which is twice as heavy, O^{16}, may consist of four alpha particles in a tetrahedral structure differing rather little from a sphere, and it does indeed show a strong evidence of collective motion of a body with tetrahedral symmetry, as epitomized by the alpha-particle model.[2] The even-even nucleus with mass between these two, C^{12}, appears not to follow the alpha-particle model,[1] and this may be associated both with the great difference between a triangle and a sphere and with the fact that the energy of dissociation into three alpha particles is as great as the average binding-energy of a nucleon. Although in Be^8 the possible alpha-particle structure, a line or "dumbbell," is also very different from a sphere, no energy is required to dissociate it into two alpha particles; so here again, there might be a tendency for the low states to be approximated by the alpha-particle model. In either the shell model or the alpha-particle model, or indeed in a blend of the two, the expected sequence of low-energy levels is very simple. The shell-model expectation is determined by the calculations which show in (LS) coupling a series of singlets, ${}^1S_1$, ${}^1D_2$, ${}^1G_4$, and similar widely-spaced levels in (jj) coupling and so also in intermediate coupling.[1] It should, however, be mentioned that states probably not belonging to the ground configuration p^n appear in the neighboring nuclei Li^7 at 6.4 Mev and C^{12} at 7.7 Mev, and might also appear in Be^8 at a comparable energy. The expectation of possible simplicity should not prejudice one, but does add interest to the experimental investigation of Be^8.

The evidence concerning the spectrum of low states in Be^8 remains conflicting. Some experiments suggest quite a number of low excited states, and other experiments, particularly those in which it has been possible to collect the most convincing statistical evidence, show only the well-known broad excited state at about 3 Mev. It is unfortunate that there are intrinsic difficulties which make it very time-consuming to collect adequate numbers of counts per point in some of the work showing the "extra" states, for the demonstration that certain other reactions do not show these states does not disprove their existence. It is always possible that an adverse matrix element is suppressing a given transition, but in the absence of some general selection rule, it becomes unlikely that this should happen at a variety of bombarding energies and angles, because a special selection that might apply for a given state of the compound nucleus or at a given angle would not be expected to apply at others. The present work is confined to reactions wherein it is not very difficult to obtain many counts per point with relatively good resolution. We have tried to vary the experimental conditions to exclude the possibility that some chance cancellation in the matrix element is hiding some of the states from us.

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