# A SEARCH FOR DOUBLE BETA DECAY IN Ca<sup>48\*</sup>

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The subject of this paper is an attempt to detect experimentally double  $\beta$  decay in Ca<sup>48</sup>. The apparatus, procedure, and control tests used in the experimental work are described. The writers have reached the conclusion that if double  $\beta$  decay occurs in Ca<sup>48</sup> its half-life is not smaller than  $0.7 \times 10^{19}$  years.

## 1. INTRODUCTION

#### Т

LHE search for double  $\beta$  decay is an amazing example of a fantastic succession of periods of hope and of disillusionment. Two times in the course of a single decade this phenomenon has been "discovered," and both times the discovery has been found to be erroneous. The history of the question is still not complete; the phenomenon has not been observed experimentally, and the succession of journal articles in recent years only gradually sets larger and larger lower limits on the lifetime of a nucleus capable of double  $\beta$  decay. In the present research we have again not succeeded in observing the event, but the limit on the half-life of the process has been raised to about  $0.7 \times 10^{19}$  years, and further steps in this direction (if indeed they are worth while) will depend on achieving considerable increases in the amount of material subjected to study.

The search for the phenomenon has been stimulated by the well known discussions<sup>1,2</sup> of the relation of the value of the period for double  $\beta$  decay with arguments in favor of or against the existence of neutrino and antineutrino as two different particles. Increase of the experimental limit of the decay period to the values now established has been regarded as strong evidence in favor of the existence of physically distinguishable neutrinos and antineutrinos.

Our views on this matter have been much changed, however, by the discovery that the law of conservation of parity is violated in the weak interactions. The result has been to give to the problem as a whole an incomparably more complex and involved character. Let us go into this in more detail.

A number of experiments performed in 1957 in the field of  $\beta$  decay have given evidence in favor

of the concept of the neutrino as a longitudinallypolarized particle. Furthermore, in the two-component scheme for the neutrino, the neutrino and the antineutrino have different orientations of the spin relative to the momentum; the antineutrino, i.e., the particle emitted along with an electron in  $\beta$ decay, has its spin and momentum parallel, while the neutrino, emitted along with a positron, has them antiparallel. This picture corresponds to the idea of 100 percent polarization of the neutrino and the law of conservation of leptonic charge. So far as one can judge, it is not in contradiction with the available experimental facts, but also it does not follow from them as an automatic consequence - the measurements are still not sufficiently accurate. In this picture there is no place for such double processes as the "neutrinoless" double  $\beta$ decay; if before the decay the atomic system contained no light particles with one sign of the leptonic charge, and after the decay such particles have appeared (for example, two electrons), then along with these particles there must appear light particles with the opposite sign of this charge (two antineutrinos).

If, however, the law of the conservation of leptonic charge is violated,<sup>3</sup> then the process of neutrinoless decay is possible owing to the occurrence of mixed types of interaction. More precisely: part of the decays in the virtual first phase of the process can go through the scalar-tensor interaction with emission of a neutrino of one type, and part through the axial vector interaction with emission of a neutrino of the opposite type. During the second phase of the process these neutrinos can be absorbed, and the process as a whole will not be accompanied by neutrino emission. It turns out<sup>4</sup> that the process of such a decay with the mixed interaction appears only when forbidden transitions are involved; this brings about a reduction of the decay probability by four orders of magnitude as compared with previous estimates.<sup>5</sup>

<sup>\*</sup>Preliminary results of this work were presented at the All-Union Conference on Nuclear Spectroscopy in January, 1957.



FIG. 1. Sketch of arrangement of measuring head. 1, 2) Scintillation counters; 3) shaft of motor for shifting of motor specimens; 4) specimens.

It must be noted that in case the two-component theory is incorrect (the longitudinal polarization of the neutrino is not 100 percent) the forbiddenness noted above can be weakened. The accuracy of the present experimental data in measurements of the longitudinal polarization of the electrons is to within 10 or 20 percent. If we assume (as is, however, not very probable) that the degree of polarization of  $\beta$  particles is 10 percent smaller than the maximum possible, we find that the probability of neutrinoless  $\beta$  decay is decreased by only two orders of magnitude instead of four.<sup>4</sup>

As for the leptonic charge, on this question there are practically no experimental data. Of the work done so far, that of Davis<sup>6</sup> might provide some information, but it is as yet unclear how it is to be interpreted. In principle, experiments on double  $\beta$  decay can also give information on the conservation of leptonic charge. It must be remembered, however, that the theory of the process involves one quantity that cannot be checked experimentally, namely the nuclear matrix element, for which one can only estimate an upper limit.<sup>5</sup>

Thus the situation in both experiment and theory indicates that continuation of experiments on double  $\beta$  decay can be of some interest.

## 2. THE PREPARATION OF THE SPECIMENS

The test and control specimens were prepared from calcium obtained by electromagnetic separation of the isotopes. To lower the proportion of extraneous atoms in the specimens, we used calcium fluoride as the chemical compound for their preparation. The test specimen, enriched in the isotope  $Ca^{48}$  to abundance 76.2 percent, contained 423 mg of  $Ca^{48}$ . The control specimen was enriched in the isotope  $Ca^{44}$  (94.7 %) and was equivalent to the test specimen in the number of molecules of calcium fluoride.

The two specimens were prepared by pressing powdered calcium fluoride and were in the shape of thin disks of diameter 37 mm. The binder was a 1 percent solution of agar in water. The disks were fixed in annular aluminum holders, with their sides covered with aluminum foil of thickness  $30 \mu$ . All the materials used in the preparation of the specimens were subjected to careful chemical analysis, and were checked for the absence of radioactivity. The results of the analyses showed that the foreign impurities in the specimens did not exceed 0.02%.

#### 3. DESCRIPTION OF THE APPARATUS

The apparatus consisted of the measuring head, completely immersed in a shielding liquid scintillation counter, electronic registering equipment, and a unit for automatic control and monitoring. The apparatus had to operate for many hundreds of hours without interruption, and was designed for the registration of extremely rare events. Therefore the greatest care was taken to assure the stability and reliability of its functioning. These considerations determined the choice of the photomultipliers and the circuits and components of the electronic equipment, and also the arrangement of the automatic control system.

The arrangement of the measuring head is shown schematically in Fig. 1. The two scintillation counters, of diameter 50 mm, are placed facing each other in a sealed casing. Between the counters there is a steel frame, in which the specimens are fixed. The frame can be shifted by means of a reversible motor connected to the control unit, so as to place between the scintillation counters either the specimen enriched with  $Ca^{48}$  or that enriched with  $Ca^{44}$ . Springs included in the mechanism for shifting the frame assure that it always takes one of the extreme positions, and that the positions of the counters relative to the scintillation counters are established reproducibly.

The distance between the median plane of the specimen and the surface of the scintillator is 1.35 mm. The solid angle over which each of the scintillation counters "saw" the specimen was  $0.464 \times 4\pi$ . The method for calculating the solid angles between the disks is described in detail in reference 7.

Each liquid scintillation counter is a 70-liter tank with polished aluminum walls, into which 21 type FEU-19M photomultipliers are directed at all angles. The tank is filled with a solution of terphenyl in toluol; the concentration of terphenyl is 3 g/l. All of the photomultipliers of the tank, which will hereafter be called "shielding" photomultipliers, were operated at the same amplification coefficient.



FIG. 2. Block diagram of the electronic part of the apparatus. 1) Cathode followers; 2) and 5) amplifiers; 3, 7, and 9) shaping units; 4) selection unit; 6) and 8) anticoincidence circuits; 10) control unit; 11) cathode-ray oscilloscope. a) To motor; b) Film advance; c) To motor; d) Time marker; e) (check)<sub>1</sub>; f) (check)<sub>2</sub>; g) Tank; h) From tank; i) Film advance; j) Time marker.

A block diagram of the electronic part of the apparatus is shown in Fig. 2. The total pulse from all the shielding photomultipliers was sent through the amplifier 5 into an electronic system, where after passing through the anticoincidence circuit 6 it was transformed by the shaping unit 7 to a rectangular pulse. The pulses coming from the photomultipliers of the measuring head were sent through the cathode followers 1 to the linear amplifiers 2 with strong negative inverse coupling. In the shaping units 3 they were transformed into pulses with flat tops and slanting trailing edges, and then went to the deflecting plates  $Y_1$  and  $Y_2$  of the twobeam cathode-ray oscilloscope 11. The formation of these "working" pulses occurred only in cases in which they were not accompanied by pulses from the screening photomultipliers.

Registration of the working pulses also occurred when quite definite conditions were fulfilled, namely when: (a) they were in coincidence to within  $0.2\mu$ sec, (b) their combined amplitude was above a prescribed value, and (c) there were no pulses from the screening photomultipliers during the time interval from 1  $\mu$  sec before the appearance of the "working" pulse to its termination. The first two conditions were imposed by the selection unit 4, and the last by the delayed anticoincidence circuit 8. If all these conditions were satisfied the shaping unit 9 was actuated; this developed a positive rectangular pulse to modulate the brightness of the oscilloscope beams. Since no alternating voltage was applied to the x axis of the oscilloscope, the pulses on the screen took the form of vertical lines directed toward each other and marked at both ends by points. A photographic attachment



FIG. 3. Specimen of the film. 1) Control pulses; 2) mark indicating the beginning of measurements on the specimen of  $Ca^{48}$ ; 3) coincident pulses registered in the course of an hour; 4) mark indicating the beginning of measurements on the specimen of  $Ca^{44}$ .

recorded this image on 35 mm film.

The anticoincidence circuit 6 shut off the pulses from the screening photomultipliers during the exposure. After each exposure the control unit 10 casued the film to be advanced by 1.5 mm.

In addition to advancing the film the control and monitoring unit performed the following functions:

(a) At the end of each hour it actuated the motor that accomplished the interchange of the specimens in the space between the photomultipliers of the measuring head.

(b) During the shifting of the frame carrying the specimens it sent out two series of standard pulses from a highly stable generator to check the stability and functioning of the electronic circuits;

(c) It actuated a device which marked the film with a conventional sign showing which specimen was in place for observation.

Figure 3 shows a reproduction of a piece of the film and gives the interpretation of all the elements of the registration. The amplitudes of the pulses registered on the film were measured by means of a projector. The measured spectrum of amplitudes was subjected to appropriate recalculation to obtain the energy spectrum.

### 4. THE SCINTILLATION COUNTERS. THE CALI-BRATION

The liquid scintillation counter prevented registration by the measuring head of ionizing particles incident on the apparatus from the space outside, and considerably decreased the probability of the registration of neutral particles and photons. The lower the energy threshold of the shielding counter,



FIG. 4. Attachment of the plastic scintillator to the FEU-12 photoelectric cathode. 1) Photoelectric cathode; 2) scintillator; 3) brass ring; 4) rubber ring; 5) cement; 6) diffusion oil; 7) threaded brass fitting; 8) hole for injection needle.

the more effectively it should fulfill its protective function. It must be taken into account, however, that lowering of the threshold is accompanied by a rapid rise of the number of counts coming from the shielding photomultipliers, owing both to the registration of weak scintillations and also to the registration of its own noise pulses in the low-energy region. An increase of the number of shielding pulses would be accompanied by a lowering of the effective time for registration and an increase of the dead time of the system. The problem of choosing the permissible counting rate can be dealt with experimentally and will be considered later on.

The choice of the electric supplies to the dynodes made it possible to improve considerably the parameters of the FEU-19M photomultipliers owing to a sharp increase of the signal-to-noise ratio. More exactly, while keeping the voltage across the divider and the value of the discrimination threshold unchanged, we were able to raise the gain of the photomultipliers by a factor of several times, with a negligible increase of the number of noise pulses.

As for the choice of the liquid for filling the tank, it is desirable to have a scintillator with the greatest light yield and with sufficient transparency for its own radiation. From this point of view the most effective substance was a solution of terphenyl in toluol, to which was added a small amount of  $\alpha$ -naphthyl phenyl oxazole ( $\alpha$  NPO) or diphenyloxazolylbenzol (POPOP). We remark that to get good results one must use toluol purified by double distillation. Toluol is a very good solvent for many organic substances, and a liquid scintillator is extremely sensitive to impurities of any kind. Therefore all of the many gaskets were made either of lead or of fluorine plastic.

Tests of the liquid scintillation counter showed that with an energy threshold for  $\beta$ -ray quanta of 90 to 100 kev the number of noise pulses did not exceed 1000 to 1500 per minute. These figures were regarded as satisfactory.

For the measuring head one must have extremely stable photomultipliers with good amplitude resolution and capable of operating for many hundreds of hours without changes in their parameters. These requirements are satisfied by the type FEU-12 multipliers developed by Vil'dgrube.<sup>8,9</sup>

For the scintillators in the measuring head we used a solid solution of tetraphenyl butadiene in polystyrene, of concentration 16 g/l. Figure 4 shows schematically the construction of the attachments used to assure good optical contact between the photoelectric cathodes of the multipliers and the plastic scintillators.

The photoelectric cathodes of the multipliers in the measuring head were connected to the body of the apparatus, and the high voltage of positive polarity was applied through the collectors. The suitability of such connections was ascertained by tests on prolonged operation of the photomultipliers.

The energy calibration of the scintillation counters for the measuring head was carried out with the conversion line of  $Ba^{137}$  (0.625 Mev) and with the position of the maximum caused by cosmic-ray mesons of minimum ionization. The calibration of the point corresponding to the internal-conversion electrons of Ba<sup>137</sup> was obtained in the following way. A small drop of a solution of a salt of radioactive Cs<sup>137</sup> was placed on a thin organic film  $(0.3\mu)$ . After the water had evaporated from the drop, the film was mounted between two rings of organic glass in such a way that the dry residue from the drop, containing the  $Cs^{137}$ , was in the center. The source was placed on an aluminum disk 3 mm thick, which in turn was fastened against the working surface of the scintillation counter. The spectrum was taken twice with a single-channel differential amplitude analyzer - once with a hole in the aluminum disk toward the source, and the other time with a solid disk. In the latter case the  $\gamma$ -ray spectrum was taken. The difference of the two spectra gave the spectrum of the electrons, with its maximum at 0.625 Mev. The energy resolution of the two scintillation counters for 0.625 Mev electrons is 28 percent (the width of the peak at half its height). On the basis of this one draws the conclusion that for 4 Mev electrons one can expect that the half-value width for the resolution is 11 percent.

The second calibration point in the region of higher energies was obtained by an independent method. The apparatus was placed so that the working counters were one above the other, and below the lower one was placed a third (auxiliary) scintillation counter, connected for coincidences with the top one (Fig. 5). The oscilloscope with the photographic attachment registered only the pulses from the working counters, and this only in the case of coincidence of the counts from the top counter and the auxiliary counter. After the film was developed attention was given only to the DOBROKHOTOV, LAZARENKO, and LUK'YANOV



cases in which pulses were registered from both working counters. This triple-coincidence method selected the vertical component of the cosmic radiation, incident normally on the working surfaces of the scintillators. After the spectrum had been constructed and the energy lost by minimum-ionization  $\mu$  mesons in the polystyrene<sup>10,11</sup> had been computed, and the correction had been applied for the difference of the light yields for  $\mu$  mesons and electrons,<sup>12</sup> a second calibration point was obtained in the neighborhood of 3.5 Mev. The Landau curve constructed on the basis of these measurements is shown in Fig. 6.

The linearity of the scintillation counters was checked by means of a  $\beta$ -ray spectrometer calibrated with the conversion line of Ba<sup>137</sup>. This gave a number of points up to energy 2 Mev; to within 2.5 percent these fell on the straight line



FIG. 6.  $\mu$ -meson spectrum obtained in calibration of the scintillation counters.





connecting the two reference points obtained with  $Ba^{137}$  and the Landau curve. The calibration curve for the scintillation counters of the measuring head is shown in Fig. 7.

#### 5. THE CONTROL EXPERIMENTS

1. The electron-counting efficiency of the measuring-head scintillation counters for counting electrons was compared with the efficiency of end-window Geiger counters with window thickness 4 mg/  $cm^2$ . It was found that in all cases the counting rate when the scintillation counter was used was 8 percent higher than with the Geiger counter. Consequently the counters used in our apparatus had an efficiency for counting electrons not less than that of a Geiger counter, whose efficiency is known to be close to 100 percent.

2. The choice of the discrimination threshold in the liquid scintillation counter circuit was made experimentally. For this purpose a curve was obtained showing the dependence of the counting rate of the number of coincident pulses in the measuring head (for constant threshold of the total energy) on the counting rate of the liquid scintillator. The results of these measurements are shown in Fig. 8. The shape of the curve shows that when the number of inhibiting pulses is larger than  $2 \times 10^5$  per minute



FIG. 8. Dependence of the counting rate of coincident pulses from the measuring head on the counting rate of the liquid scintillator.



FIG. 9. Integrated spectrum of coincident pulses: 1) on the surface; 2) underground at a depth of 65 m water equivalent.

the shielding by anticoincidences becomes less effective owing to the increase of the dead time of the anticoincidence circuit, whereas for a number of inhibiting pulses less than  $4 \times 10^4$  per minute the shielding effectiveness is still insufficient. The presence of a plateau indicates that there is adequate shielding effectiveness against most kinds of external radiation. In view of all this, the discrimination threshold of the liquid scintillation counter circuit, operated under these circumstances, was chosen at about  $10^5$  counts per minute. We estimate that the corresponding energy threshold is about 0.1 Mev for  $\gamma$  radiation.

3. A check on the identity of the calibration of the two counters in the measuring head was carried out under the following conditions. All the units of the apparatus were fully actuated, and photographic registration of the energy spectrum was obtained for 12 hours. To shorten the time of the measurement the tests were carried out not under the conditions of the experiment, under the ground, but on the surface. The energy spectra were compared with each other; besides this, comparisons were made between the sums of the amplitude values of the pulses for each of the channels. The total energy of all the pulses registered by one of the counters of the measuring head was 8959 Mev, that for the other counter, 8983 Mev. The good agreement of both the spectra and the amplitude totals indicates that there is identity of the calibrations of the two counters.

4. The integrated energy spectrum of the sum of the coincident pulses was taken both on the surface and underground. These tests were to determine the effect of the operating conditions on the background, i.e., on the counting rate of coincident pulses. The measurements were carried out with the measuring head not shielded by the liquid scintillator. The results are shown in Fig. 9.

The comparison of the counting rates for total energy 3 Mev shows that burying the apparatus under the ground leads to a decrease of the background by a factor of 23. Thus it is quite expedient



FIG. 10. Energy spectra of background of two sets of apparatus for measuring double  $\beta$  decay: 1) 1956; 2) 1957;  $\Delta E = 0.2$  Mev.

to make the measurements underground.

5. The effects of shielding by anticoincidences and of screening with lead were strikingly demonstrated in the following experiment. The completely assembled apparatus was set for registration of coincidences with the threshold at a total energy of 1 Mev. Numbers of coincidences in 1 hour were compared with the anticoincidence circuit turned on and with this circuit turned off; in the former case 17 coincidences were recorded, in the latter case 56. After this the apparatus was surrounded on all sides with a layer of lead 10 cm thick, and the measurement was repeated with the anticoincidence circuit turned on. Seven coincidences were registered during two hours. Thus the use of the lead shield leads to a further lowering of the background of coincidence counts by a factor of 5.

The control experiments that have been described confirmed the correctness of the calibration of the apparatus, provided a reasonable choice of the discrimination threshold, and demonstrated the effectiveness of the protections against background counts that were used in the experiment. This last point is illustrated by Fig. 10, which shows the energy spectra of the background for the apparatuses for measuring double  $\beta$  decay used by the writers in 1956<sup>13\*</sup> and in the present work. It must be remarked, however, that the apparatus was relatively poorly shielded against neutron radiation.

### 6. THE MEASUREMENTS AND THE PROCESSING OF THE RESULTS

The conditions under which the measurements were made are, properly speaking, clear from the

<sup>\*</sup>In Fig. 2 of reference 13, through an oversight of the editors, the energy scale is displaced to the right by 0.5 Mev.

E, Mev	Ca48		Ca"		
	Series 1	Series 2	Series 1	Series 2	
$3-4 \\ 4-5 \\ 5-6 \\ 6-7$	$\begin{array}{c} 1,16{\pm}0.52\\ 1.16{\pm}0.52\\ 0.92{\pm}0.47\\ 0.23{\pm}0.23 \end{array}$	$\begin{array}{c} 1.00 \pm 0.58 \\ 0.67 \pm 0.47 \\ 1.00 \pm 0.58 \\ 0.33 \pm 0.33 \end{array}$	$\begin{array}{c} 0.93 \pm 0.47 \\ 0.93 \pm 0.47 \\ 0.23 \pm 0.23 \\ 0.47 \pm 0.33 \end{array}$	$2.00\pm0.82$ 0.00 0.33\pm0.33 0.33\pm0.33	

 TABLE I. Comparison of spectra from first and second series

 of measurements

information given above. The measurements were carried out under ground at a depth of 65 m water equivalent, and the apparatus was shielded against the effects of the external background by the anticoincidence circuit and by a lead screen. Two series of measurements were carried out, the first in December 1956 — January 1957, and the second in July — August 1957. The time of measurement with each of the specimens was 430 hours in the first series and 300 hours in the second.

The stability of the parameters of the apparatus during the time of the measurements was checked automatically. The constancy of the operating conditions of the counters in the measuring head was checked regularly by the values of the voltage and current in the circuit of the VS-9 high-voltage rectifier that supplied the counters, and also by the constancy of the counts of both coincident and single pulses coming from the measuring head.

The measurements provided films on which were registered the amplitude distributions of the coincident pulses that came in the course of the experiment from the photomultipliers of the measuring head. After the amplitudes had been measured the spectra obtained were subjected to statistical analysis. For this purpose the whole time of the measurements was divided into equal intervals, and for each of the photomultipliers the numbers of pulses of amplitude exceeding a prescribed value that arrived within each interval were counted. The two series of numbers so obtained were analyzed both from the point of view of agreement with the Poisson distribution, and as to whether they agreed with each other. The results of the analysis showed that no crude errors came in in the process of handling the photographic films, verified the identical functioning of the two scintillation counters, and showed the stability of the operation of the apparatus as a whole.

As has already been mentioned, two series of measurements were made. For each of the series the spectra of the total energies of the coincident pulses were constructed, and a comparison of these spectra was made. The results of the comparison are shown in Table I. This table gives the counting rates per 100 hours of observation for the energy intervals indicated.

As can be seen from these data, the agreement of the spectra is quite satisfactory. After the processing of the results had shown that the laws of statistics are obeyed and that the spectra of the two series are in agreement with each other, these spectra were combined into the total spectra shown in Fig. 11

#### 7. DISCUSSION OF THE RESULTS; CONCLUSIONS

To interpret the obtained results we have first to find out in what energy interval we must compare the spectra for Ca<sup>48</sup> and Ca<sup>44</sup>. The energy interval in which the effect of double  $\beta$  decay is to be sought depends on the expected energy of the decay, the assumptions about the process, and the energies lost by the electrons in the material of the specimens used. Let us examine this problem, starting from the assumption of the "neutrinoless" decay process, in which the entire energy released in the decay is divided between the two electrons.

For the case of Ca<sup>48</sup> the energy of the decay is known with adequate accuracy from mass-spectrographic data,<sup>14</sup> and has the value  $4.3 \pm 0.1$  Mev. The average total energy loss of the electrons that emerge from the material of the specimen in such



FIG. 11. Spectra of total energies of coincident pulses obtained in 730 hours of measurement with each of the specimens: solid line –  $Ca^{46}$ ; dashed line –  $Ca^{44}$ ;  $\Delta E_2 = 0.2$  Mev.

a way that they go into different counters was determined by computation. For the specimens used in the present experiment it is 0.4 Mev, and corresponds to the energy loss of a relativistic electron in a path length equal to twice the thickness of the specimen. The computation was made on the assumption of absence of angular correlation between the decay electrons. According to this the search for the effect should be made in the region of 3.9 Mev.

The width of the energy interval in which one should compare the spectra of  $Ca^{48}$  and  $Ca^{44}$  depends both on the errors of the apparatus and also on a possible "smearing out" of the spectrum of the total energy of the electrons from double  $\beta$  decay. The probable errors introduced by the apparatus are the following:

1. The spread in amplitude introduced by each scintillation counter  $(\pm 5.6 \text{ percent})$ .

2. The error in the energy calibration of the scintillation counters  $(\pm 3 \text{ percent})$ .

3. The lack of stability of the gain coefficient of the apparatus  $(\pm 1.5 \text{ percent})$ .

4. Errors in measurements on the film  $(\pm 1.5 \text{ percent})$ .

Thus, taking account of the fact that two counters were functioning simultaneously, we get for the root-mean-square error of the apparatus  $\pm 9$  percent, which corresponds to a spread of  $\pm 0.35$  Mev in the energy. To be on the safe side we increase this figure to 0.5 Mev.

The effect of "smearing out" of the spectrum, owing to the spread of the amount of energy lost by the electrons in the material of the specimen, should lead to a shift of the maximum of the spectrum toward lower energies. This has been partly taken into account in the introduction of the correction for the loss of energy in the material of the specimen, and the part not so included can scarcely exceed 0.4 Mev.

Thus the spectra of the total energy of the electrons is to be analyzed in the region from 3.0 to 4.4 Mev. As can be seen from Fig. 11, in this energy range during the 730 hours there were registered 11 cases of coincidences when the specimen enriched in Ca<sup>48</sup> was between the scintillation counters, and 12 cases for the specimen of Ca<sup>44</sup>. The difference "Ca<sup>48</sup> - Ca<sup>44</sup>" consequently is  $(-1 \pm 4.8)/730$  counts per hour, or  $(-0.14 \pm 0.66)$  counts per 100 hours.

The half-life was determined from the formula

$$\tau = \ln 2 \frac{N_0}{A} \frac{km\eta}{\Delta n}$$

where m is the amount of the substance to be stud-

ied contained in the specimen (in grams); A is the mass number;  $\Delta n$  is the difference of the numbers of counts per unit time from the working and control specimens; k is a factor corresponding to the geometrical effectiveness for coincidence counting, on the assumption of isotropic distribution of the decay electrons;  $\eta$  is a coefficient characterizing the "transparency" of the specimen for the decay electrons; and  $\,N_{0}\,$  is Avogadro's number. The coefficient  $\eta$  was calculated on the assumption of no angular correlation between the decay electrons and for the energy distribution function of these electrons given in reference 5; it was found to be 0.52. The value of the coefficient k, as determined on the basis of the value of the solid angle over which each of the counters "saw" the specimen, was 0.43. After inserting the numerical values of the quantities one gets the following numerical formula for the half-value period:

$$\tau = \frac{0.9}{\Delta n} \cdot 10^{19} \text{ years,}$$

where  $\Delta n$  is per hundred hours.

Following the established tradition, we shall give here the value of the half-life calculated on the assumption of no angular correlation between the decay electrons. Inclusion of the theoretically expected angular correlation<sup>15</sup> would lead to a decrease of both the figures given in the present paper and of those in other papers on searches for double  $\beta$  decay by about a factor of three.

The extremely small intensity of the count in the energy range in question, and the consequent high probability of considerable statistical fluctuations, lead to the danger of obtaining false conclusions if one regards as significant differences values of  $\Delta n$  greater than just a single standard error  $\sigma$ . Only results exceeding at least twice the standard error can be given any weight. An analysis of the spectra obtained here shows that there is not a single energy interval of reasonable width in the range from 2.4 to 7.0 Mev in which  $\Delta n$  would exceed  $2\sigma$ . Therefore we can determine only a lower limit to the half-life for double  $\beta$  decay of Ca<sup>48</sup>. Using the numerical formula and inserting instead of  $\Delta n$  the quantity  $2\sigma$ , we find that the half-life for double decay of Ca<sup>48</sup> is not less than  $0.7 \cdot 10^{19}$  years.

Thus the result of our measurements is a negative one. Clearly a further reduction of the background could not bring any change in the situation, because a colossal length of time is needed for reliable measurements with a small quantity of the material. Thus there remains only one way to make further progress — increasing the actual

Year	Reference	Result	Method	
1955 1956	McCarthy <sup>16</sup> The writers <sup>13</sup>	$1.1 \times 10^{17}$ years (+) > 1 × 10 <sup>18</sup> years	Scintillation "	counters
1956	Awschalom <sup>17</sup>	$> 2 \times 10^{18}$ years	n	"
1957	Present paper	$\geq$ 0.7 × 10 <sup>19</sup> years	"	"

TABLE II

mass of the isotopes studied. If besides this one is able to increase the dimensions of the counters, then the need will arise for a further decrease of the background. This last may be possible only after a detailed study of the factors determining the shape of the energy spectrum of the background. This problem is not considered in the present paper.

Let us sum up. In Table II are collected the results of the experiments made to search for double  $\beta$  decay in Ca<sup>48</sup>. In this table the sign "plus" indicates a positive result of the search.

The lower limit now obtained on the half-value period of the double decay is larger by almost an order of magnitude than the value corresponding to incomplete polarization of the neutrino. Therefore it can be asserted that the experimental result of our study agrees with the two-component theory of the neutrino, in which the degree of polarization is 100 percent. The possibility of a two-component scheme with occurrence of double  $\beta$  decay through forbidden transitions (with nonconservation of the leptonic charge) remains open.

The necessary reservations limiting the certainty of the conclusions we have drawn, owing to lack of accurate knowledge of the nuclear matrix element and the degree of angular correlation of the electrons, have already been pointed out in the text.

The writers take occasion to express their sincere appreciation to I. S. Shapiro for an interesting and fruitful discussion of the problem of double  $\beta$ decay. We are also deeply grateful to I. V. Galkin for building the electronic part of our apparatus and to K. S. Mikhailov for the syntheses and the preparation of the scintillators. <sup>3</sup>M. G. Mayer and V. L. Telegdi, Phys. Rev. **107**, 1445 (1957).

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