quadrupole angular distributions although we are not in disagreement with the $l=4$ contributions, predicted by the shell model.[24] In addition, the sum of their differential cross sections equals that of the first 2+ in Cr^{52}. As seen in Table III their $B(E2)\downarrow$ values are not equal to the Cr^{52} state.

It is interesting to note that the sum rule predicted by the weak-coupling model[25] holds even though the coupling is not weak and none of the other predictions are realized.

ACKNOWLEDGMENTS

The authors would like to thank F. S. Goulding, D. L. Landis, and L. B. Robinson for the excellent electronic and computing systems which they developed and helped us to use, and Claude Ellsworth for preparing the targets. The entire staff and crew of the 88-in. cyclotron must be acknowledged as coauthors of this paper, for without their efforts and cooperation it would not have been written.

[24] H. O. Funston, N. R. Robertson, and E. Rost, Phys. Rev. **134**, B117 (1964).
[25] A. de-Shalit, Phys. Rev. **122**, 1530 (1961).

PHYSICAL REVIEW VOLUME 146, NUMBER 3 17 JUNE 1966

Limits for Lepton-Conserving and Lepton-Nonconserving Double Beta Decay in Ca^{48}†

E. der Mateosian and M. Goldhaber
Brookhaven National Laboratory, Upton, New York

(Received 10 February 1966)

A search has been made for double beta decay in Ca^{48} using a new technique which involves the use of CaF_2 crystals as scintillation detectors. These crystals, enriched in either Ca^{48} or Ca^{40}, were used to detect ionizing particles originating within the crystals. The advantages of this technique are (1) a 4π geometry for a source without self-absorption and (2) good resolution which is independent of the relative angular and energy distribution of the two electrons. A lower limit of 2×10^{20} yr is placed on the lepton-nonconserving double beta decay without neutrinos. The determination of a lower limit for the Dirac-type double beta decay (with emission of two neutrinos) is complicated by impurities in the Ca^{48} used. By resorting to the usual coincidence type of search for double beta decay, which makes the effect of the impurities negligible, we established a lower limit of 5×10^{18} yr for this process. This is comparable with the lower limit of the theoretical estimates.

INTRODUCTION

PHENOMENOLOGICALLY, double beta decay[1] is concerned with a search for two possible reactions:

$$(A,Z)\to(A,Z+2)+2e^-, \tag{1}$$

$$(A,Z)\to(A,Z+2)+2e^-+2\bar{\nu}. \tag{2}$$

It has been emphasized, especially by Pauli[2] and Greuling and Whitten,[3] that the existence or nonexistence of reaction (1) is probably the most sensitive test for lepton conservation. Both reactions are expected to take place as the result of a second-order effect due to the same nucleon-lepton (weak) interaction which gives rise to the usual single beta decay.[4] It has been pointed out,[5] however, that reaction (1) could also conceivably take place in the first order if there existed certain lepton-nonconserving interactions. Prior to 1957, when only parity-conserving interactions were considered in beta decay theory, the double beta decay process was regarded as a possible means of determining whether the Majorana or Dirac description of the neutrino was the correct one. Both of the above reactions are allowed by the Majorana theory, but only reaction (2) can take place in the Dirac theory. In the particular case of Ca^{48}, which appears to be one of the most favorable nuclei to study, these theories predicted[4] a half-life of $3\times10^{15\pm2}$ yr for reaction (1) and $1\times10^{21\pm2}$ yr for reaction (2). Further, the two reactions are distinguished by the fact that the sum of the two electron energies in reaction (1) is constant and equal to the transition energy while it varies continuously in reaction (2) up to the same energy as its limit. Many searches were made in this period for double beta decay and several early apparently positive results were later disproved except one obtained by an indirect chemical method.

† Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] G. F. Dell' Antonio and E. Fiorini, Nuovo Cimento **17**, Suppl. I, 132 (1960).
[2] W. Pauli, Nuovo Cimento **6**, 204 (1957).
[3] E. Greuling and R. C. Whitten, Ann. Phys. (N. Y.) **11**, 510 (1960).
[4] S. P. Rosen and H. Primakoff, *Alpha-, Beta- and Gamma-ray Spectroscopy* (North-Holland Publishing Company, Amsterdam, 1965), pp. 1499–1516.
[5] G. Feinberg and M. Goldhaber, Proc. Natl. Acad. Sci. **45**, 1301 (1959).

The direct, counting measurements were made with sufficient sensitivity so that lifetimes shorter than 10^{18} yr could be excluded and this led to the conclusion[6] that decay (1) did not take place. With the discovery of parity nonconservation, the Majorana theory in its simplest form had to be given up, but recent reformulations[3,7] of the theory again allow under certain assumptions for the possibility of the occurrence of reaction (1) as a second-order effect, adding further interest to the search for both reactions (1) and (2) with improved techniques.

About a dozen years ago we realized that increased sensitivity could be gained in the investigation of double beta decay by giving up coincidence counting techniques for a method which has the advantage of a 4π geometry.[8] Specifically, we looked for the double beta decay of Ca^{48} by using a large single crystal of CaF_2 as a scintillator and recording events whose origins were within the crystal. Since our source, Ca^{48}, was an integral constituent of the crystal the method offered the further advantages of the equivalent of a "thin source" and fairly good resolution independent of the relative angular or energy distribution of the two electrons.

The total available energy for double beta decay in Ca^{48} has been calculated to be 4.27 MeV from atomic mass measurements made by Giese and Benson.[9] Energies based on (pn) reaction data[10] did not agree with this value until Chasman, Jones, and Ristinen[11] showed that the $Ca^{48}(p,n)$ reaction leads to an excited state of Sc^{48}, 131 keV above the ground state. The addition of this transition energy to the Q value of the reaction made the reaction data consistent with the mass spectroscopic data.[12] Single beta decay of Ca^{48} is energetically possible to the ground state and to two known excited states of Sc^{48} but has not been observed.[13] These transitions are highly forbidden because of the large difference of spin between the ground state of Ca^{48} and the levels in Sc^{48}, as shown in Fig. 1. In neutrino-less double beta decay the two electrons would share the total energy difference between Ca^{48} and Ti^{48}.

Our earliest attempt to detect double beta decay in Ca^{48} was made with a large single crystal of pure

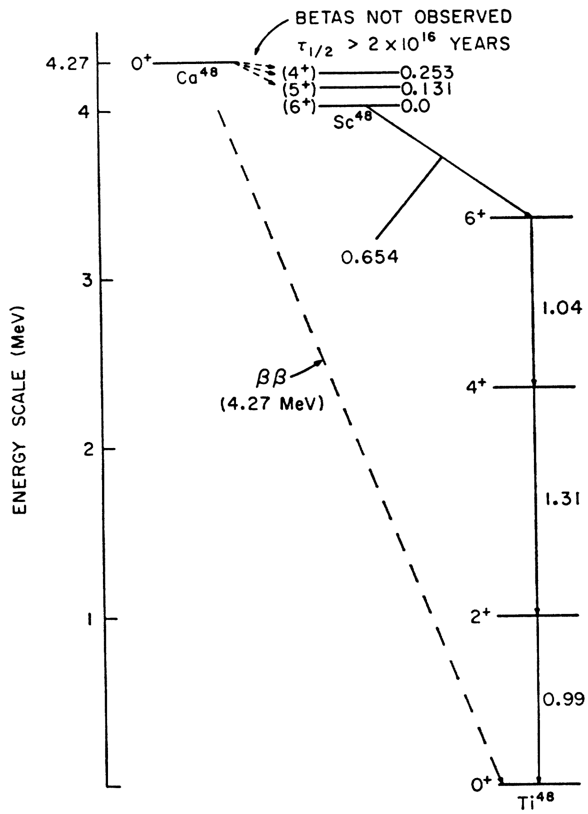

FIG. 1. Energy diagram of $A=48$ mass chain.

unenriched CaF_2 borrowed from the Harshaw Chemical Company. This crystal was a right cylinder 4.0 in. high, 4.5 in. in diameter and 2951 g in weight. Figure 2 shows the pulse-height spectrum obtained with this crystal and gives details of the geometry of the experiment in the insert. The total counting rate in the 4.27-MeV energy region as delimited by the resolution of the crystal for electrons leads to a lower limit of 2.5×10^{16} yr for the half-life for neutrino-less double beta decay. A conservative statistical treatment of our data, estimating the background by extrapolation, enabled us to set a lower limit of 10^{17} yr, similar to the values obtained at that time by other authors.[14,15]

NEUTRINO-LESS DOUBLE BETA DECAY

At our request the Isotope Separation Department at Oak Ridge National Laboratory produced sufficient enriched Ca^{48}, assaying 96.59%, to enable us to have a crystal of CaF_2 grown[16] containing about 10.6 g of Ca^{48}. The performance of this crystal as a scintillator was improved over that of pure CaF_2 by the addition of $\sim$0.5% Eu as an activator. Crystals of both normal

[6] J. S. Allen, *The Neutrino* (Princeton University Press, Princeton, New Jersey, 1958), Chap. 6.
[7] K. M. Case, Phys. Rev. **107**, 307 (1957).
[8] Preliminary reports of these results have been given as follows: M. Goldhaber, Bull. Am. Phys. Soc. **8**, 46 (1963); E. der Mateosian and M. Goldhaber, in *Proceedings of the 12th Annual International Conference on High-Energy Physics, Dubna, 1964* (Atomizdat, Moscow, 1965); Bull. Am. Phys. Soc. **9**, 717 (1964); and Proceedings of the Neutrino Conference Cern, Geneva, 1965 (unpublished).
[9] C. F. Giese and J. L. Benson, Phys. Rev. **110**, 712 (1958).
[10] L. A. König, J. H. E. Mattauch, and A. H. Wapstra, Nucl. Phys. **31**, 18 (1962) [C. M. Johnson (private communication)].
[11] C. Chasman, K. W. Jones, and R. A. Ristinen, Phys. Rev. **140**, B212 (1965).
[12] The latest results are included in the 1964 Atomic Mass Table given by J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, in Nucl. Phys. **67**, 1 (1965), from which one obtains an energy of 4.267 MeV for the double beta process in Ca^{48}.
[13] J. W. Jones and T. P. Kohman, Phys. Rev. **85**, 941 (1952).
[14] J. A. McCarthy, Phys. Rev. **97**, 1234 (1955).
[15] M. Awschalom, Phys. Rev. **101**, 1041 (1956).
[16] All crystals were grown by the Harshaw Chemical Company, Cleveland, Ohio.

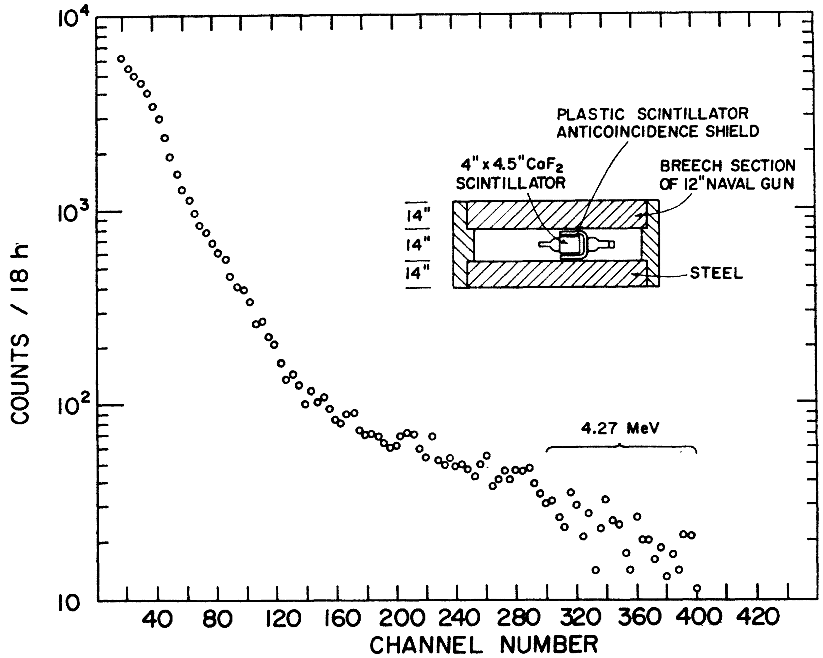

FIG. 2. Search for double beta decay of Ca^{48} in single crystal of normal CaF_2. Spectrum of counts seen with a 4-in. by 4.5-in. diam. normal CaF_2 crystal used as a scintillator to detect pulses originating within the crystal. Neutrino-less double beta decay would yield a peak at 4.27 MeV.

CaF_2 and $Ca^{40}F_2$ were also grown under identical conditions and these were used as controls. Figure 3 shows the spectrum observed with the crystal enriched in Ca^{48}. The very definite peaks in the region from channels 40 to 100 were due to either electron- or alpha-particle-emitting impurities. The light output of this crystal for electrons and alpha particles was measured and was found to be four times larger for an electron of a given energy (in the MeV region) than for an alpha particle of the same energy. Hence, the peak in channel 100 represents either a 2-MeV electron or an 8-MeV alpha particle. Strong sources of homogeneous 2-MeV electrons are exceedingly rare, but a homogeneous alpha-particle group of 7.68 MeV is emitted by Po^{214}, which is a member of the uranium series of radioactive elements. An impurity of 1 part per million of uranium would be sufficient to give the observed peak in channel 100, and a suitable mixture of thorium and uranium impurities

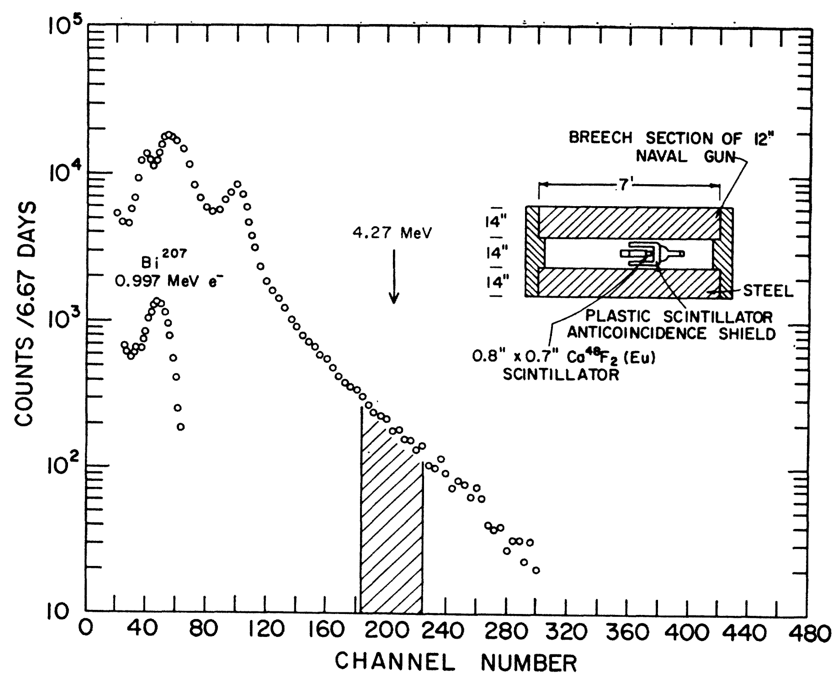

FIG. 3. Spectrum of counts seen with a crystal of $Ca^{48}F_2$ (Eu) enriched to 96.59% Ca^{48}. An external source of Bi^{207} conversion electrons was used for calibration.

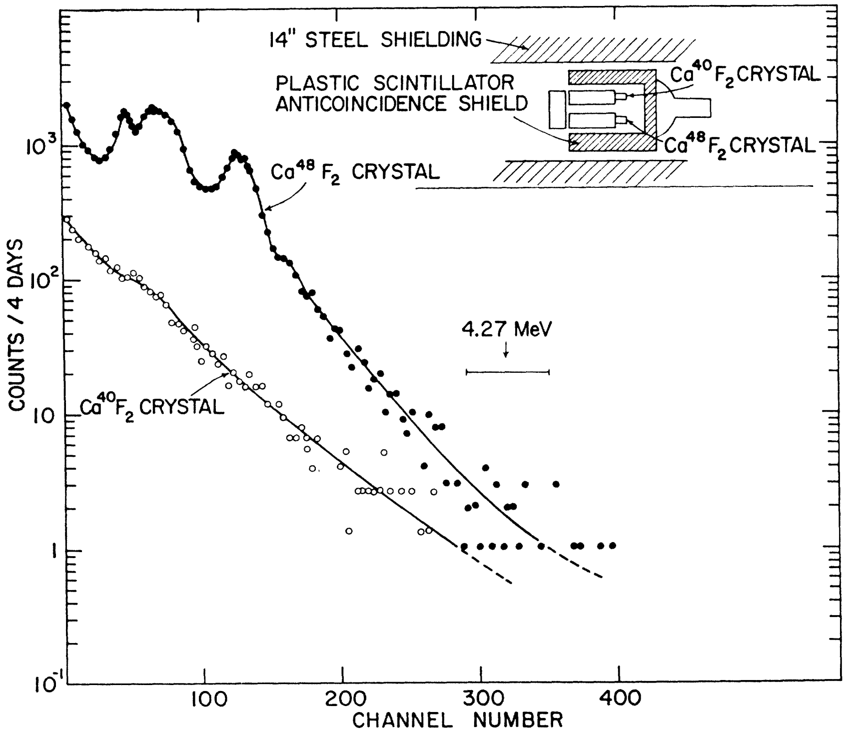

FIG. 4. Spectrum of counts seen with a crystal of $Ca^{48}F_2$ (Eu) enriched to 96.59% Ca^{48}. This run differs from that of Fig. 3 in that the Ca^{48} was repurified and a new crystal was grown. The reduced counting rate can be judged by referring to the scales of each figure. A run with a control crystal of $Ca^{40}F_2$ (Eu) is also shown for comparison.

could account for the main features of the spectrum observed.

The Ca^{48} was sent back to Oak Ridge for purification and a second crystal was grown which contained 11.4 g of Ca^{48}. Again control crystals of normal CaF_2 and $Ca^{40}F_2$ were grown. Figure 4 shows that a lower counting rate was obtained with this crystal, as a comparison of the scales of Figs. 3 and 4 will verify. In the region where neutrino-less double beta decay would be observed (4.27 MeV) the counting rate is low and close to a background count obtained with a similar crystal of $Ca^{40}F_2$. After subtracting this background, we find that the total remaining counts represent a lower limit of 1.2×10^{19} yr for the double-beta-decay half-life. Experimental details of the run are again shown in an insert in Fig. 4. In this run data were accumulated simultaneously with both the $Ca^{48}F_2$ and the $Ca^{40}F_2$ detectors, which were placed side by side within a plastic scintillator operated in anticoincidence with both detectors. The entire assembly was housed within a section of a naval gun with 14-in.-thick walls of steel.

Five such runs were made totaling 28.7 days of running time. The difference of counts of the combined runs in the Ca^{48} and Ca^{40} crystals is plotted against channel number in Fig. 5.

In neutrino-less double beta decay one would expect to see a peak at 4.27 MeV (channel 140 in Fig. 5) with a resolution which may be deduced from the resolutions of either the impurity peaks in the Ca^{48} spectra or externally impressed internal conversion electron peaks. A conservative estimate of the lower limit for neutrino-less double beta decay was made by adding all the counts in a 0.6 MeV wide region centered around 4.27 MeV. A lower limit of 2.4×10^{19} yr is found either by adding the experimentally obtained counts or by taking the total counts under the fitted smooth curve. A somewhat better lower limit may be established by considering the fluctuation of the experimental points about the smooth curve, representing the extrapolated background. The average deviation of the points about the line may be calculated and twice this deviation may be considered to be the minimum deviation a point would

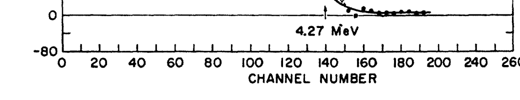

FIG. 5. Difference of counts in Ca^{48} and Ca^{40} crystals. The dashed curve labeled 1×10^{20} yr indicates the deviation from the smooth curve that would have been noticed if neutrino-less double beta decay took place in Ca^{48} with $\tau_{1/2}=1\times10^{20}$ yr. The larger dashed peak labeled 7×10^{18} yr represents neutrino-less double beta decay with a lifetime corresponding to a previously published lower limit. (See Ref. 17.) Actually, this peak should be about three times larger. (See text and Ref. 17.)

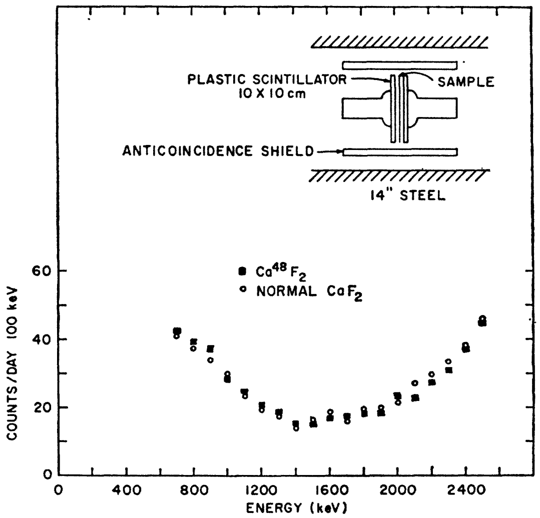

FIG. 6. Spectra of the combined energies of coincidence pulses for Ca^{48} and for normal Ca.

need in order to be significantly different from the line. Then, barring the accidental presence of a dip in the background spectrum at 4.27 MeV, the smallest double-beta-decay peak which would be detectable could be considered to be one that is at least equivalent to twice the average deviation. This criterion gives a lower limit of 2.3×10^{20} yr. Still a third criterion, which has been used by previous investigators, is to take the statistical accuracy with which the total counts in the region under investigation are known. If twice the standard deviation is taken one obtains $\tau_{1/2}>1.8\times10^{20}$ yr. For comparison, in Fig. 5 a peak corresponding to 1×10^{20} yr is superimposed on the background (dashed line). Therefore, 2×10^{20} yr appears to be a reasonable lower limit. A larger peak corresponding to the previous lower limit of Dobrokhotov *et al.*[17] is also shown for comparison.

DOUBLE BETA DECAY WITH TWO NEUTRINOS

Because of the impurities in the Ca^{48} we were not able to obtain a significant lower limit for the Dirac type of double beta decay (two electrons plus two neutrinos), which would yield a continuum ending at 4.27 MeV. To avoid the interference of the alpha-particle emitting impurities we had the Ca^{48} powdered and packaged in a moderately thin layer (0.162 g/cm^2) so that it could be used as an external source for coincidence counting. By adopting this more generally used technique for investigating double beta decay we lost the advantage of sensitivity but benefitted by avoiding the background from the radioactive impurities, since the α particles no longer constituted a measurable portion of the radiations that were able to emerge from the sample and be counted in the detectors, and the single beta rays did not contribute directly to the coincidence counting rate. The detectors were two plastic scintillators with 100 cm^2 of counting area which were mounted within a plastic scintillator anticoincidence shield in the 14-in.-thick steel shield. A sample of normal CaF_2 was used as a control. Similar spectra were observed with a Ca^{48} source and the normal Ca source. Two runs were made, one for 4 days, the other for 6 days, and in one run the Ca^{48} "spectrum" had more counts than the normal Ca "spectrum" in the region of concern while the reverse was true in the second run. These fluctuations were not statistically significant. The two runs were combined and are plotted in Fig. 6. Again experimental details of the counting arrangements are given in an insert. The spectra shown were obtained by demanding coincidences between the two detectors which were not accompanied by counts in the anticoincidence shield, adding the outputs of the two detectors and accumulating these in a multichannel pulse-height analyzer. The spectrum observed with the Ca^{48} sample in place is, within statistics, identical to the spectrum with the normal Ca sample in place. The difference between the spectrum found for Ca^{48} and normal Ca is plotted in Fig. 7 where it is compared with the theo-

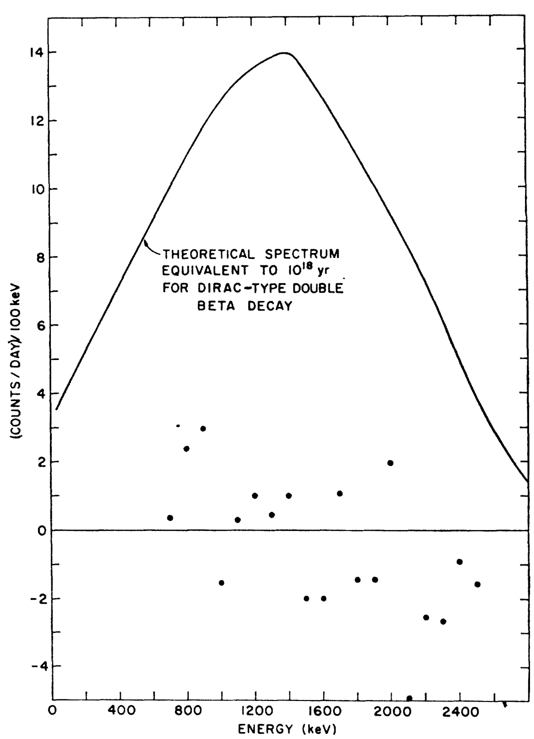

FIG. 7. Difference of the coincidence spectra for Ca^{48} and normal Ca of Fig. 6. A theoretical shape for the spectrum of double beta decay with neutrinos is shown which is equivalent to a half-life of 10^{18} yr. This curve is shifted 400 keV to the left to account for self-absorption of the electrons in the sample.

[17] E. I. Dobrokhotov, V. R. Lazarenko, and S. Yu Luk'yanov, Zh. Eksperim. i Teor. Fiz. 36, 76 (1959) [English transl.: Soviet Phys.—JETP 9, 54 (1959)]. {Since the preparation of this paper, new results have been reported by the last two authors of this reference. See V. R. Lazarenko and S. Yu. Luk'yanov, Zh. Eksperim. i Teor. Fiz. 49, 751 (1965) [English transl.: Soviet Phys.—JETP 22, 521 (1966)]}.

TABLE I. Theoretical predictions and experimental limits for double beta decay of Ca^{48}.

Nature of lifetime determination	Half-life (years) Neutrino-less	Two neutrinos (Dirac type)	Authors
Theory	$3\times10^{15\pm2}$	$1\times10^{21\pm2}$	Rosen and Primakoff[a]
		$>10^{19}$	Meichsner[b]
		10^{19}	Beliaev and Zakharev[c]
		$>2\times10^{19\pm1}$	Greuling and Whitten[d]
Experiment	$>2\times10^{18}$ [e]		Awschalom[f]
	$>7\times10^{18}$ [e]		Dobrokhotov, Lazarenko, and Luk'yanov[g]
	$>5\times10^{19}$ [e]	$>3\times10^{18}$	Lazarenko and Luk'yanov[g]
	$>4\times10^{18}$	$>4\times10^{18}$	Shapiro, Frankel, Koicki, Wales, and Wood[h] (improved values by private communication)
	$>2>10^{20}$	$>5\times10^{18}$ yr	Present work

[a] See Ref. 4.
[b] See Ref. 19.
[c] See Ref. 20.
[d] See Ref. 3.
[e] These values are a factor 3 too high due to the expected angular correlation between the electrons. See Refs. 17 and 18.
[f] See Ref. 15.
[g] See Ref. 17.
[h] See Ref. 21.

retically expected curve[3] equivalent to a half-life of 10^{18} yr. A quantitative estimate for the lower limit was made by taking the total number of counts under the Ca^{48} curve, correcting for the fact that the instrument has a low-energy cutoff at 700 keV, taking twice the statistical fluctuation of this number as the measure of the lower limit and correcting for the coincidence geometry. This yields $\tau_{1/2}>5\times10^{18}$ yr.

CONCLUSION

In Table I[18–21] we have summarized some of the significant theoretical estimates[3,4,19,20] and experimental observations[15,17,21] concerning the double beta decay lifetime of Ca^{48}. The "e" following some of the experimental values listed for the lower limit to the half-life calls attention to a fact pointed out by Dobrokhotov *et al.*[17] that those lifetime values which were obtained by coincidence measurements ought to be reduced by a factor of 3 because of the angular correlation[18] expected between the two electrons in the neutrino-less double beta emission.

The search for double beta decay in Ca^{48} has still not led to the observation of this phenomenon. One can see from Table I that the experimentally determined lower limit for the half-life for the Dirac-type (two-neutrino) double beta decay is just approaching the more optimistic theoretical estimates. This higher order effect must be expected to exist, but it is difficult to predict its half-life since the matrix elements involved are hard to estimate. It has been reported[22] that the possible existence of double beta decay has been detected in Te^{130} by an indirect chemical method.

Our results on the neutrino-less double beta decay can be considered an improved test of lepton nonconservation. If lepton-nonconserving interactions exist[5] which allow double beta decay as a first-order process, our results indicate that the coupling strength of such interactions must be more than 10^{14} times weaker than the Fermi coupling. Thus, the continued search for neutrino-less double beta decay has helped to establish the law of lepton conservation on essentially as firm a basis as charge conservation and baryon conservation.

ACKNOWLEDGMENTS

The authors wish to thank Dr. George L. Rogosa of the U. S. Atomic Energy Commission for his help in obtaining separated isotopes of Ca. L. O. Love, Supervisor of the Electromagnetic Isotope Separation Department and members of the Chemistry Group at Oak Ridge National Laboratory, especially H. R. Gwinn and R. L. Bailey, have been very cooperative and helpful in the preparation and purification of the Ca^{48} isotope. Finally, the authors wish to express their appreciation of the very extensive cooperation of the Harshaw Chemical Company, especially, E. C. Stewart and Dr. C. F. Swinehart, who tackled the difficult task of growing the scintillation crystals of enriched Ca isotopes without loss of material.

[18] H. Primakoff, Phys. Rev. **85**, 888 (1952).
[19] L. Meichsner, Phys. Rev. **117**, 489 (1960); **120**, 552 (1960).
[20] V. B. Beliaev and B. N. Zakharev, Zh. Eksperim. i Teor. Fiz. **34**, 505 (1958) [English transl.: Soviet Phys.—JETP **7**, 347 (1958)].
[21] M. H. Shapiro, S. Frankel, S. Koicki, W. Wales, and G. T. Wood, Bull. Am. Phys. Soc. **10**, 424 (1965). The values in the table were given in an invited paper presented by M. H. Shapiro at the Summer Meeting of the American Physical Society, New York, 1965, and in private communication.
[22] R. J. Hayden and M. G. Inghram, Natl. Bur. Std. Circ. No. **522**, 189 (1953); N. Takaoka and J. Okano, Shitsuryo Bunseki **12**, 195 (1965).