features of the resulting hyperfine structure are that the center of gravity of a given line does not change significantly and that two small satellites appear, one on either side of the main line. Their intensity is about 1/100 of that of the main line and hence they were observed only for the four most intense lines.

The B_0 values agree most closely with those given in reference 3. The value of the quadrupole coupling constants are slightly smaller than those previously reported, and the corresponding coupling constants in the 1st excited bending mode are smaller than those in the ground state by about 1 part in 250. The value of the l-type doubling constant compared to that in the $J=2\rightarrow3$ transition shows the slight J dependence previously reported for OCS and HCN.[21]

I wish to thank Professor Robert Beringer for his very helpful advice and criticism throughout the course of this work.

[21] R. G. Shulman and C. H. Townes, Phys. Rev. **77**, 421 (1950).

PHYSICAL REVIEW VOLUME 86, NUMBER 4 MAY 15, 1952

Neutrino Recoils Following the Capture of Orbital Electrons in A^{37}*

GEORGE W. RODEBACK† AND JAMES S. ALLEN
Department of Physics, University of Illinois, Urbana, Illinois
(Received January 21, 1952)

The energy spectrum of the recoil nuclei resulting from orbital electron capture in A^{37} has been determined by a time-of-flight measurement of the recoil ions. A^{37} disintegrations occurred within a gas-filled source volume, where the total pressure was approximately 10^{-5} mm of Hg. The time-of-flight distribution, as measured by a multichannel delayed-coincidence technique, has a definite maximum time limit. Furthermore, the shape and extent of the distribution correspond to that expected from monoenergetic recoils, originating in an extended source and having an energy of 9.7 ev as predicted by the single neutrino hypothesis. Zero and short time coincidences were observed in addition to the above distribution. Further measurements indicate these to be the result of electron scattering associated with A^{37} decays not originating in the source volume.

I. INTRODUCTION

THE measurement of the energy of the recoil nucleus associated with orbital electron capture in an isotope in gaseous form should provide an answer to the question of whether or not single neutrinos are emitted in this type of radioactive decay, as was pointed out by Crane.[1] This experiment was an attempt at such a measurement in which radioactive A^{37} was used.

The isotope A^{37} has properties which make it well suited for a recoil energy determination by a time-of-flight measurement. First of all, the expected recoil velocity results in a time-of-flight of the order of a few microseconds for a drift distance of several centimeters. In addition, the excited atom, following orbital electron capture, returns to its ground state primarily by the emission of one or more Auger electrons of less than 3000-ev energy. The detection of these low energy electrons with an electron multiplier provides a means of initiating the time measurement. Since the low velocity recoil atoms which emit the Auger electrons are either singly or multiply ionized, they can be accelerated through an electric field to an energy which renders them easily detectable with an electron multiplier.

The primary experimental requirements for this measurement were to continuously maintain a gaseous source of constant and suitable strength and to record the data in a reliable fashion for long periods of time. The geometry finally employed proved to be a compromise between good velocity resolution and high coincidence counting rates.

II. PROPERTIES OF A^{37}

Assuming the emission of a single neutrino, the orbital-electron capture disintegration of A^{37} is represented by the equation

$$A^{37}+e_{K,L}\rightarrow Al^{37}+\nu+Q,$$

where $e_{K,L}$ is the captured orbital electron, ν the emitted neutrino, and Q is the disintegration energy. If the neutrino has zero rest mass, the Q of the above reaction is given by the $A^{37}-Cl^{37}$ mass difference. A value of 816 ± 4 kev for this mass difference has been obtained from a recent $Cl^{37}(p,n)A^{37}$ threshold measurement[2] together with a n-p mass difference[3] of 782 ± 1 kev. If the entire disintegration energy is carried away

* This investigation was supported jointly by the AEC and ONR.

† Now at North American Aviation Company, Downey, California. The material in this paper was submitted by G.W.R. to the University of Illinois in partial fulfillment of the requirements for the Doctor of Philosophy degree.

[1] H. R. Crane, Revs. Modern Phys. **20**, 295 (1948).

[2] Richards, Smith, and Browne, Phys. Rev. **80**, 524 (1950).

[3] Taschek, Jarvis, Argo, and Hemmendinger, Phys. Rev. **75**, 1268 (1949).

by the single neutrino and the recoiling nucleus and if we assume that linear momentum is conserved between the neutrino and the recoiling nucleus, the energy of the nuclear recoil should be 9.67 ± 0.08 ev corresponding to a velocity of 0.711 ± 0.004 cm/μsec. The contribution of the binding energy of the orbital electron to the reaction energy is negligible, and has been omitted in the above computations.

A^{37} decays entirely by orbital electron capture with a half-life of 34 days.[4] About 93 percent of the disintegrations result from K capture with about 90 percent of this fraction resulting in emission of Auger electrons; the remainder is K x-ray emission. The other 7 percent of the total number of disintegrations result from L capture as was first reported by Pontecorvo *et al.*[5] Using the proportional counter technique, this group measured the spectrum of energies due to the emission of Auger electrons and x-rays[6] and confirmed the existence of Auger electron energies of about 2400, 2600, and 200 ev corresponding to $K-L^2$, $K-LM$, and $L-M^2$ converted electrons. These energies are in agreement with the values computed from the known critical absorption wavelengths of chlorine.[7]

According to Morrison and Schiff,[8] about 0.05 percent of the disintegrations should result from radiative orbital electron capture. In this radiative capture process the available energy is shared between a neutrino, a γ-quantum, and the recoiling nucleus. An almost continuous spread of recoil momenta should result from this type of disintegration. However, this effect was not observed in the present experiment since the expected counting rate was much smaller than the chance coincidence rate. Maeder and Preiswerk[9] have recently shown that radiative electron capture does occur in Fe^{55}.

III. RECOIL CHAMBER AND METHOD OF RECORDING DATA

Figure 1 shows a schematic cross section of the chamber in which the time-of-flight measurements were made. During the run the total pressure of the gases in the chamber, including that of the A^{37}, was maintained at about 10^{-5} mm Hg. This corresponds to a mean free path of about 500 cm for argon atoms. The shaded trapizoidal cross section indicates the effective source volume which was defined by baffles and also by the region seen simultaneously by both detectors. The necessary baffles, shields, and wire gauze grids (denoted by dotted lines) were all maintained at ground potential with the exception of grid 3 which was maintained at approximately -4500 volts with respect to ground. A

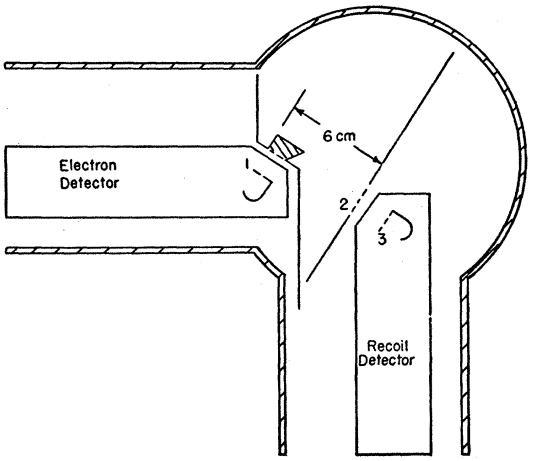

FIG. 1. Schematic of A^{37} time-of-flight apparatus. The effective source volume is indicated by the shaded trapezoidal cross section in front of the grid 1. The recoil Cl^{37} ions resulting from a disintegration within the source volume traverse a field free path to grid 2 and then enter the ion counter after an acceleration through a potential difference of 4.5 kv.

delayed coincidence was recorded for an A^{37} disintegration occurring within the source volume when the resulting Auger electron passed through grid 1 into the electron detector and when, in addition, the ionized chlorine atom traversed the field free distance to grid 2 and was counted by the recoil detector. Both detectors were Allen-type electron multipliers.[10]

Delayed coincidences of the recoil detector output with respect to the electron detector output were recorded by a 20-channel delayed coincidence circuit. The outputs of the two multipliers were fed through two identical channels consisting of preamplifiers, linear pulse amplifiers, and discriminators. The final output of the electron detector channel emerged from a pulse shaper circuit and initiated the delayed coincidence circuit. The final output of the recoil detector channel also was shaped and fed into the multichannel coincidence circuit. Time calibration of the entire delayed-coincidence circuit, including measurement of resolving times and total delays for individual channels, was accomplished by the introduction at the preamplifier inputs of two pulses separated by a precisely known time interval.

The 20 delayed coincidence channels follow consecutively in time and usually the adjacent channels overlap by as much as 10 percent of the time width of an individual channel. During the period of a run the total counts for each channel are recorded and chance counts for each channel are computed on the basis of its resolving time and the measured singles counts from both detectors. The true counts are given by the difference between the total coincidence counts and the calculated chance counts. What is plotted, however, is the number of true counts per channel of unit time

[4] Weimer, Kurbatov, and Pool, Phys. Rev. **66**, 209 (1944).
[5] Pontecorvo, Kirkwood, and Hanna, Phys. Rev. **75**, 982 (1949).
[6] Kirkwood, Pontecorvo, and Hanna, Phys. Rev. **74**, 497 (1948).
[7] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), p. 792.
[8] P. Morrison and L. I. Schiff, Phys. Rev. **58**, 24 (1940).
[9] D. Maeder and P. Preiswerk, Phys. Rev. **84**, 595 (1951).
[10] J. S. Allen, Rev. Sci. Instr. **18**, 739 (1947).

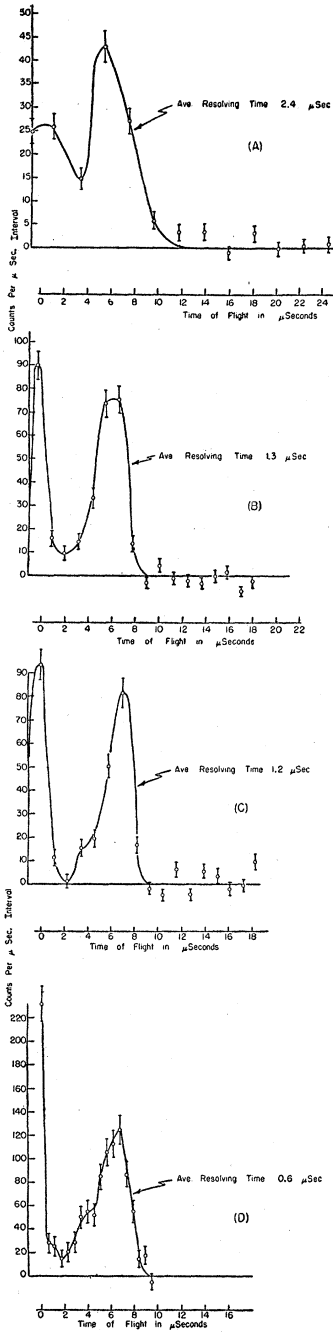

FIG. 2. Time-of-flight distributions obtained with a geometry similar to that of Fig. 1. The mean time-of-flight path was 5 cm. The average channel width of the time recorder was progressively decreased in order to yield increasingly greater details of the distribution.

width (i.e., true counts divided by resolving time) as a function of the total elapsed time to the midpoint of a coincidence channel. The plot then represents an experimental determination of the differential time distribution of the recoils coming from the source volume.

The relative statistical accuracy in the number of true counts can be defined as the ratio of true counts to the probable statistical error in the observed number of counts. An expression for this ratio in terms of the constants of the counting arrangement can be obtained. Assuming an extended source which emits monoenergetic recoils, it can be shown that this ratio is proportional to $(\tau T)^{\frac{1}{2}}$ where τ is the resolving time of a channel and T is the total elapsed counting time. Therefore, when the details of the time of flight spectrum are desired (implying that the resolving time be shorter than the time spread expected for the recoils coming from the extended source volume), increased resolution is obtained at the expense of the statistical accuracy, and this loss can be compensated for only by increasing the counting time. For most of the experiments to be described below, the source strength was adjusted to produce a true to chance ratio greater than unity. The statistical accuracy in the number of coincidences per channel then was determined by the length of the observation period.

IV. RESULTS

Figure 2 shows several time-of-flight distributions obtained for a geometry similar to that of Fig. 1 with a mean traversal distance for the recoil ions of about 5 cm. The conditions for each run were similar, except that the resolving times of the coincidence channels were successively decreased, thus yielding increasingly greater detail of the distribution. The statistical accuracy was made nearly the same for each plot by controlling the time duration of each run.

The peak at about 7 μsec is the result of the recoils originating in the source volume. The abrupt cessation of the time-of-flight distribution at about 9 μsec, with no further coincidences indicated out to at least 35 μsec, indicates the absence of recoils with velocities less than the expected value of 0.711 cm/μsec. This sharp cutoff, together with the fact that the distribution has the general shape expected for monoenergetic recoils from the source volume, is interpreted as experimental verification of the unique energy of the recoil atoms. This explanation assumes that the zero and short time counts of the distribution can be satisfactorily explained.

Following the runs shown in Fig. 2, the apparatus was altered slightly to give a better defined source volume. Figure 3 shows the resulting distribution. The dashed curve is the predicted distribution based on the shape and location of the source volume and the assumed value of recoil velocity which gives the best fit.

The far edge of the measured distribution was used to calculate an experimental value for the recoil velocity based on the time for a singly ionized Cl^{37} atom to traverse the maximum field-free distance from the source volume to grid 2 of Fig. 1. The following considerations entered into the calculation of the delay time to be used:

1. The ideal time-of-flight distribution is modified

due to the thermal velocities of the A^{37} atoms in the source. The root-mean-square velocity at room temperature is about 0.04 cm/μsec and results in a spreading at the base of the ideal distribution. The expected spread is very similar to the observed far edge of the distribution in Fig. 3. Therefore, the intercept of the dotted curve at 7.8 μsec is taken as the observed time-of-flight corresponding to the maximum recoil distance. However, further corrections must be made to this value based on the following:

2. The time required for a singly ionized Cl^{37} atom to be accelerated between grids 2 and 3 (Fig. 1) and thence to travel at constant velocity to the sensitive region of the first dynode is calculated to be 0.3 μsec.

3. The field between grids 2 and 3 (Fig. 1) was found to have penetrated the supposedly field-free region between the source volume and grid 2. This field distribution was subsequently measured, and calculations based on this show that the traversal time in this region for a singly ionized atom is decreased by 1.0 μsec. For doubly ionized atoms this is 1.4 μsec. With the above corrections for a singly ionized atom the corrected maximum time is $7.8-0.3+1.0=8.5$ μsec. The maximum distance is 6.0 cm, resulting in a recoil velocity determination of 0.71×0.06 cm/μsec, which is in excellent agreement with the expected value of 0.711×0.004 cm/μsec.

The following two additional corrections are negligible compared to the uncertainties of this experiment:

1. Time of flight of Auger electrons from source volume to electron detector.

2. The momentum of the recoiling nucleus is equal in magnitude and opposite in direction to the vector sum of the momenta of the neutrino and the Auger electron, since the electron emission occurs before the recoil has moved through an appreciable distance. When the recoil atom and electron are at 90° as in the present arrangement, the maximum change expected in the recoil momentum is 0.2 percent.

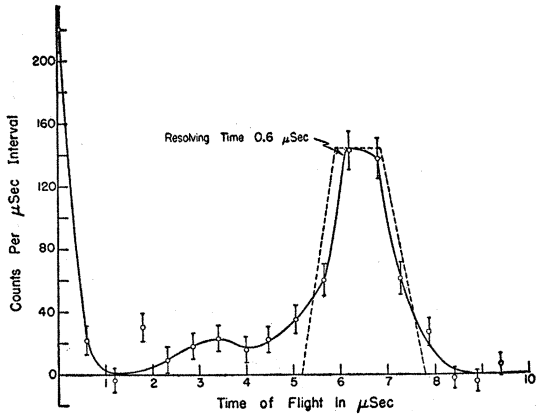

FIG. 3. Time-of-flight distribution obtained with improved definition of the source volume. The dashed curve is the distribution expected for monoenergetic recoils coming from the source volume. The tail of the solid curve in the region of 8 μsec is due to the thermal velocities of the A^{37} atoms in the gaseous source.

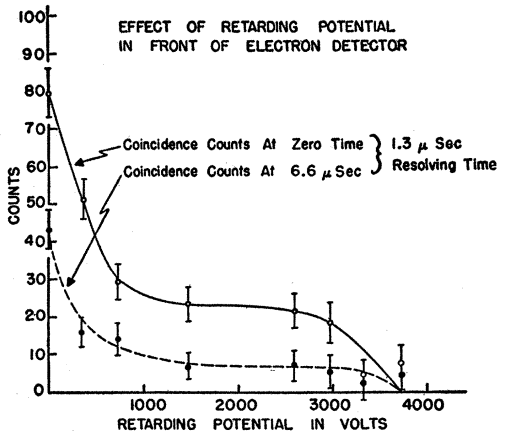

FIG. 4. Curves showing the effect of retarding potentials on the electrons entering the electron counter.

Based on the geometry and estimated counting efficiencies, the probability of an A^{37} disintegration in the source volume resulting in a coincidence count is about 10^{-5}. Calculations indicate that during the above run the specific activity in the source volume was about 200 disintegrations per sec per cm^3.

By imposing a retarding potential on the left side of grid 2 in Fig. 1 and noting the effect on the time-of-flight distribution as this potential is varied, it should be possible to determine the relative degrees of ionization of the recoil atoms. Due to instrumental difficulties it was possible only to verify that a retarding potential of 16.5 volts eliminated the distribution of delayed counts from the source volume.

V. ORIGIN OF ZERO AND SHORT TIME COUNTS

A series of measurements were made in an attempt to explain the origin of the zero time peak and the other coincidences occurring at times shorter than those of the main time-of-flight peak. Zero time coincidences were recorded for two separate runs made under identical conditions except that the first was made with no ion accelerating field in front of the ion detector (first dynode at ground) while the second run was made under the normal conditions for a time-of-flight measurement. It was found that there were 20 percent as many zero time coincidences without the accelerating field as with it. Therefore, at least 80 percent of the zero time coincidences are to be associated with the presence of the 4500-volt difference of potential between grids 2 and 3. It is likely that some of the coincidences measured without the field can be detected only under this condition and may be due to x-ray electron or electron-electron coincidences resulting from cascade Auger processes in the Cl^{37} atom.

A series of runs were made to determine the effect of an electron retarding potential in front of the electron detector. Grid 1 together with the first dynode of the electron detector (Fig. 1) was maintained at a negative potential with respect to the grounded source volume

baffle. Figure 4 shows the results of these runs where the zero time and 6.6-μsec (at maximum of delayed peak) coincidences are plotted as a function of the retarding potential at the electron detector. For each of these runs the total number of ion detector counts was maintained at the same value.

On the basis of the above results the following conclusions are made concerning a normal run:

1. The electrons counted by the electron detector for the zero time and delayed coincidences have nearly identical energy spectra.

2. The delayed coincidences at 6.6 μsec, when the electron retarding potential is greater than 2800 volts, probably are due to x-ray ion coincidences originating in the source volume.

3. No appreciable number of zero time coincidences originate in the source volume. Probably the best evidence for this is the fact that as mentioned in Sec. IV a retarding potential of 16.5 volts eliminated the delayed coincidences at 6.6 μsec. This indicates the absence of high energy positive ions which would be necessary for an essentially zero time of flight over a path of 5 cm.

4. Most of the zero and short time counts are believed to be due to disintegrations occurring in or near the region between grids 2 and 3. The coincidence would be between the recoil ion and an Auger electron which undergoes an elastic scattering process with the baffle structure surrounding the source volume and subsequently enters the electron detector. The acceleration of electrons by the field between grids 2 and 3 will aid this process. This is indicated by Fig. 4, where appreciable zero time counts are recorded for electron energies above the maximum Auger electron energy.

5. A few zero time counts may originate from those secondary electrons emitted at the first dynode of the ion detector which escape outwards and after acceleration between grids 3 and 2 are elastically scattered into the electron detector.

6. According to the above explanations a number of delayed coincidences involving scattered Auger electrons are to be expected from disintegrations occurring in the region between the source volume and grid 2. From solid angle considerations a peak in the number of coincidences should occur near the source volume, and going toward grid 2, this number should at first fall to a minimum and finally increase rapidly as grid 2 is approached. The general shape of the time of flight distribution (Fig. 3) below 5 μsec seems to agree with these predictions.

VI. METHOD OF PROVIDING SOURCE

The A^{37} used for these measurements was initially obtained from the Oak Ridge reactor, where it was prepared by neutron bombardment of Ca^{40}. The radioisotope arrived mixed with a small amount of air. Preceding a run, the oxygen and nitrogen were removed from approximately 1 millicurie of the 30 millicurie source by exposure to outgassed metallic calcium heated to 565°C. The remaining gas was then introduced into the high pressure side of a three-stage diffusion and booster pump combination after the fore pump had been sealed off from the system. The resulting pressure was a few microns of Hg. The calcium purifier was now operated at 300°C and adequately performed the functions of the usual fore pump and, in addition, removed the impurities from the A^{37}. With this arrangement the recoil chamber could be kept evacuated to less than 10^{-6} mm of Hg.

During a run the A^{37} in the reservoir at the rough vacuum side of the pumps was allowed to leak into the recoil chamber through a needle valve at a rate giving a suitable counting rate for the experiment. A steady state was very quickly reached and the recycling process could be steadily maintained for over twelve hours. The best source used resulted in a total pressure of about 10^{-5} mm of Hg in the recoil chamber. A relative measure of the source strength present in the reservoir was continuously provided by a monitor. This monitor was a thin window mica Geiger counter which was exposed to the gaseous source and responded to the small percentage of Cl^{37} recoils emitting K radiation. The high absorptivity of charcoal at liquid air temperatures was utilized for storage of the A^{37} between runs. All but a very small fraction of the source in the reservoir could be collected and sealed off in a cooled tube containing outgassed charcoal powder. This made it possible to use the same source sample many times.

VII. CONCLUSIONS

The results of this experiment indicate that for most of the A^{37} orbital electron capture disintegrations, the missing energy of a disintegration is shared between the recoil nucleus and a single neutrino. Linear momentum is shown to be conserved between the recoil nucleus and a single neutrino. Additional experiments will be necessary to further clarify the origin of the zero time and short time coincidence counts, and further refinements in the time of flight method should yield a more accurate value of the recoil velocity. Future investigations based on the techniques of this experiment should reveal the details of the processes which occur when the electronic levels of the excited Cl^{37} atom return to the ground state.