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RADIOCHEMICAL AND SPECTROMETER STUDIES OF SOME NEW NUCLEAR ISOMERS PREPARED BY CYCLOTRON BOMBARDMENT

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Hirdaya Behari Mathur

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October 5, 1954

ABSTRACT

This dissertation concerns the preparation and identification of some new nuclear isomers of various nuclear types. The results are discussed in relation to the predictions of the independent particle shell model of the nucleus.

A detailed study of the decay schemes of Cs^{127} and Cs^{125} revealed branching decay to isomeric levels in the odd-mass isotopes Xe^{127} and Xe^{125}. E3 transitions of 75 seconds and 55 seconds, respectively, were observed. By bombardment of zirconium, niobium, yttrium and silver targets in the 184-inch cyclotron it was possible to produce and study the isomeric pair Nb^{89m} and Nb^{89}.

In addition to the study of odd-mass number isotopes, information was obtained on previously unreported even-mass number isomers. A detailed study of the radiations of Mo^{90} revealed the presence of the new odd-odd isomers, 24-second Nb^{90m_1} and 10-millisecond Nb^{90m_2}, in addition to the ground state, 14.6-hour Nb^{90}.

Isomeric levels in even-even nuclei are rare but in the study of 14.6-hour Nb^{90} a delayed state of 2.2-Mev decay energy has been established in the even-even nucleus Zr^{90}.

New nuclear data unrelated to isomerism is included on some isotopes of cesium, xenon and iodine.

Radiochemical and Spectrometer Studies of Some New Nuclear Isomers Prepared by Cyclotron Bombardment

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I. INTRODUCTION

The experimental work described in this thesis has to do with the preparation, identification and study of new examples of nuclear isomerism. Because the results corroborate the predictions of the nuclear shell model it is desirable in this introduction to discuss briefly the present status of the subject of nuclear isomerism and the application of the shell model to it.

The first case of nuclear isomerism was discovered by Hahn[1] in 1921 when he found the radioactive substance UZ which was isotopic and isobaric with the well-known UX_2, but which differed from it with respect to half life and the radiations emitted.

This example of isomerism remained unique for many years, but with the discovery of artificial radioactivity in 1934, and especially the wide enlargement of the field by the production of many new radioactive substances by cyclotron and pile bombardments, the possibility of investigating new examples of nuclear isomerism arose. The first established case was that of Br^{80} investigated by Kurchatov *et al.*[2] and Fermi and co-workers.[3] In two recent reviews of isomerism[4,5] the list of known cases had grown to a total of from 100 to 200.

The first attempt to provide a theoretical basis for isomerism was made by von Weizsäcker[6] who first suggested that the nuclear isomeric states decayed into one another by gamma emission and that the long delay in this process was accounted for by large changes in angular momentum accompanying the decay. This suggestion has proved to be correct and considerable theoretical work has gone into the quantitative development of the idea. The goal of theory has been the calculation of the half life of such a transition, given the magnitude of the spin change, the mass number, the energy and the nature of the radiation, i.e., whether electric or magnetic. A relationship given by Axel and Dancoff[7] was used for several years but has recently been replaced by a more satisfactory equation provided by Weisskopf.[8,9] The Weisskopf formula for electric transitions as given by Goldhaber and Sunyar[4] is,

$$\tau_{\gamma_{sec}} = \frac{\Delta I[1.3\ldots(2\Delta I + 1)]^2}{2(\Delta I + 1)} \frac{(\Delta I + 3)^2}{3^2} \frac{1}{\rho^{2\Delta I}} \left(\frac{137}{W}\right)^{2\Delta I + 1} \frac{\hbar}{mc^2}$$

$\tau_{\gamma_{sec}}$ = the half life in seconds

ΔI = spin change

ρ = nuclear radius = $0.532A^{1/3}$

W = transition energy in units of mc^2 = E/mc^2

E = transition energy in electron volts

A = mass number.

To obtain the magnetic transition probability the above equation is supplemented by

$$(\tau_\gamma)_{\Delta I, \text{magnetic}} = (\tau_\gamma)_{\Delta I, \text{electric}} \; 5.11A^{2/3} ;$$

The importance of the Weisskopf formula has induced Montalbetti[10] to construct a nomogram of it to facilitate its application.

In addition to the emission of electromagnetic radiation it is possible for a nuclear isomer to deactivate by the process of internal conversion. An orbital electron may be ejected to carry off the energy of excitation. The competition between this process and gamma emission is expressed by the conversion coefficient which is defined as the ratio of the number of electrons so ejected to the number of quanta of electromagnetic radiation emitted. The calculation of internal conversion coefficients is capable in principle of rather exact treatment since nuclear wave functions are not involved. An extensive table of K-shell coefficients calculated by machine methods on the Mark-I calculator of the Computation Laboratory of Harvard University has been published by Rose et al.[11] These tables show values of the K-shell conversion coefficients for transition energy greater than 150 kev for the first 5 electric and magnetic multipoles. The K-shell conversion coefficients for lower gamma energies and L-shell conversion coefficients are being calculated for later publication. These tables are important because the conversion coefficient is a sensitive function of energy, multipole order and atomic number. The value of the conversion coefficient is therefore a valuable clue to the nature of an isomeric transition.

The existence of isomerism implies the occurrence of nearby nuclear energy states differing greatly in angular momentum. An adequate theory of isomerism must explain the occurrence of such states. It has been established that the spin-orbit coupling, single-particle model of Mayer[12] and of Haxel, Jensen, and Suess[13] allows predictions which agree

closely with the experimental results in a high percentage of cases, particularly for odd-A isomers.

This model predicts that isomerism should occur only in certain "islands," namely those nuclei in which the number of the odd neutron or odd proton is somewhat lower than one of the magic numbers--50, 82, or 126. Since the stability of the magic numbers is presumably due to the fact that the spin orbit coupling splits levels with large angular momenta in such a way as to lower appreciably those of high j, the levels of high spin occur just before the magic numbers in competition with the lowest spins of the previous shell. Thus, in particular, islands of isomerism in odd nuclei should occur when the odd neutron or odd proton is between 39 and 49 or between 63 and 81, or between about 100 and 125.

When the odd-neutron or odd-proton number lies between 39 and 49 the competition is between the $g_{9/2}$ and the $p_{1/2}$ orbits. The spin change of 4 and a change in parity predicts an M4 transition. Many such transitions have been observed. In addition, in this region some E3 transitions are also found but only in cases in which more than one nucleon (or less than one hole) occupies the 9/2 orbit. Kurath[14] and Talmi[15] have shown that several particles in a 9/2 shell are able to couple their spins in various ways so that instead of a j value of 9/2, a resultant of 7/2+ may be expected.

Likewise, in the second island between 63 and 81, many isomers of M4 type are observed but only in odd-neutron nuclei. The shell model predicts low-lying $h_{11/2}$, $d_{3/2}$ and $s_{1/2}$ orbits of almost equal energy. The isomeric transitions in this region, therefore, decay from the $h_{11/2}$ to the $d_{3/2}$ state, giving an M4 isomer, and in cases where the

ground state has been known to have a spin of 1/2 the isomeric transition is followed by a second gamma ray corresponding to the transition from the $d_{3/2}$ to the $s_{1/2}$ orbit. In some cases the $d_{5/2}$ orbit slips in between the $h_{11/2}$ and $d_{3/2}$ orbits, introducing thereby the possibility of an E3 isomer corresponding to the transition $h_{11/2} \rightarrow d_{5/2}$. This may be followed by an additional M1 transition from $d_{5/2} \rightarrow d_{3/2}$ or an E2 transition from $d_{5/2} \rightarrow s_{1/2}$, according as the ground state is $d_{3/2}$ or $s_{1/2}$.

The Mayer single-particle model then makes possible predictions of groups of nuclides in which isomerism may be expected and the multipole order of the radiations involved. Hence, in order to compare experimental results with the theory it is important to determine the multipole character of the radiation. This can be done in three ways.

Comparison of the half life of the transition with the predictions of the Weisskopf[8,9] lifetime-energy relationship discussed above for various multipole orders is one approach. A second approach is to determine the conversion coefficient and to compare it with the accurate values given by Rose et al.[11] for various multipole types. Experimentally this is a difficult measurement because the absolute measurement of low-energy Auger electrons or low-energy x-rays is involved. Furthermore, in the case of the measurement of x-rays the fluorescence yield must be known and for many elements this has not been accurately measured.

A third method of estimating the multipole character is the measurement of the ratio of conversion in the K shell and in the various subshells. The "K/L ratio" can be easily and accurately measured for many cases by comparing the intensity of the K and L conversion lines measured in a beta-ray spectrograph. This ratio depends upon the multipole

character, the energy and the nuclear charge Z. When the computational program of Rose and co-workers is completed, accurate values of predicted K/L ratios will be available. In the meantime since rather unreliable theoretical and nonrelativistic curves on K/L conversion ratios are available at present, Goldhaber and Sunyar[4] recommend the use of empirical curves. By plotting all available K/L ratios against Z^2/E they found that all transitions which have the same multipole order (determined by lifetime-energy relationship or conversion coefficient data) lie on the same curve and thus define the order for the particular curve. Hence in many cases it is possible to determine the spin change and parity from the measured K/L ratios.

The striking success that the single-particle model of Mayer has had in predicting isomerism and nuclear spectroscopic configurations for odd-mass number nuclides has made it a universally used basis for predictions of these and related properties in all types of nuclei. The data on odd-odd and even-even nuclei, however, are not so readily explainable.

In the case of the odd-odd nucleus, according to the rules set forth by Nordheim,[16] configurations of the odd neutron and of the odd proton are the same as in an odd-A nucleus with the same number of nucleons in the odd-particle group. There will be a coupling of neutron and proton groups and each will contribute to the resulting nuclear spin. The manner in which this grouping takes place is not very well understood at present, but the Nordheim rules indicate that spins of odd-odd nuclei are often very large. It is possible to account for isomerism in most odd-odd nuclei in which it has been observed but there is

no "systematics" of isomerism in such cases to permit much in the way of prediction.

There are only a handful of well-established examples of isomers among the even-even nuclei and these examples are Hf^{180m}, Pb^{202m}, Pb^{204m}, and Pb^{206m}. It is, therefore, not possible to draw any definite conclusions regarding the systematics of nuclear configuration in the case of even-even isomers. Goldhaber and Sunyar[4] in their article on classification of nuclear isomers made the important observation that the first excited state of even-even nuclei is 2+, the ground state being assumed to be 0+ in all cases so far known. In a recent article on the excited states of even-even nuclei G. Scharff-Goldhaber[17] has shown that out of 68 cases examined, 66 have the first excited state of spin 2 and even parity. In summarizing the observations on even-even nuclei, G. Scharff-Goldhaber has given the empirical rule that for the n^{th} excited state $I \leq 2n$. There are only a few exceptions to this rule. Hence one would not expect isomers with $\Delta I > 2$ in even-even nuclei.

In the experimental work reported in this thesis an attempt was made to find new examples of nuclear isomerism. The single-particle model was used as an aid in selecting likely nuclides for preparation and study and for interpretation of the details of the isomerism when found. The nuclides to be examined were prepared by bombardment of various targets in the 184-inch cyclotron of the Radiation Laboratory and they were purified by radiochemical separation techniques. Some new experimental techniques were developed to handle the special problems encountered, particularly in handling radioactive gases, and these are discussed in a special section.

In the case of odd-A nuclides isomerism was detected in Xe^{127}, Xe^{125}, and Nb^{89}. The results are a clear demonstration of the value of the shell model in such cases. These examples are discussed in detail in Sections II and III below.

An interesting case of isomerism in an odd-odd nucleus was found in the case of Nb^{90}. Here a cascade of two isomeric transitions involving three distinct isomeric levels was found. This work is discussed in Section IV.

As mentioned above, only a very few even-even nuclei show isomerism involving adjacent levels with spins differing by more than 2. In the present study such a transition in an even-even nucleus appears to have been measured. This is the 2.30-Mev transition in the decay of 14.6-hour Nb^{90} to Zr^{90}. This case is discussed in Section V.

During the course of the work a number of previously unreported nuclides were investigated. These nuclides were not found to exist in isomeric forms but the information on their decay properties is worth reporting. The nuclides in this group which include Xe^{121}, Xe^{122} and Xe^{123} are discussed in Section VI.

Experimental techniques are discussed in a final Section VII.

II. STUDIES OF NEUTRON-DEFICIENT ISOTOPES OF CESIUM AND OF THE E3 ISOMERS Xe^{127m} AND Xe^{125m} PRODUCED BY THEIR DECAY

This section deals with a study of nuclear isomers of odd-mass number where the isomerism is the result of an odd neutron just below the closed shell of 82 neutrons. The odd-mass isotopes of xenon are quite interesting in this respect. The Mayer model predicts that $h_{11/2}$, $s_{1/2}$

and $d_{3/2}$ states will lie close together in the neutron number region just below 82. Hence long-lived M4 transitions are to be expected, and in fact Xe^{135}_{81}, Xe^{133}_{79}, Xe^{131}_{77} and Xe^{129}_{75} show such isomerism. This situation has been thoroughly reviewed by Bergström.[18]

In the present work isomerism in Xe^{127}_{73} and Xe^{125}_{71} was studied. Here it was found that the M4 transition type had given way to an E3 type because the $d_{5/2}$ level, which in higher mass isotopes had been located higher than the $h_{11/2}$ level, had dropped below this level. The $d_{5/2}$ level is permitted by the shell model but in its present form the theory is incapable of stating whether or where the crossover of the $h_{11/2}$ and $d_{5/2}$ levels should occur.

Because the half lives of the E3 transitions are considerably shorter than those of the M4 type it was considered desirable to look for the expected isomers in the decay of cesium parent activities. Hence the neutron-deficient isotopes Cs^{127} and Cs^{125} were prepared by (α,xn) bombardment of iodine, purified radiochemically, and subjected to detailed study. Previously existing data on Cs^{127} and on Xe^{127m} were considerably extended as will be discussed next. The previously unreported activities Cs^{125} and Xe^{125m} were found. During the course of the work new information on Cs^{130} and Cs^{123} was accumulated. This provided no data on isomerism and is reported but briefly at the end of this section.

A. The Radiation of Cs^{127} and the Isomer Xe^{127m}

Fink, Reynolds and Templeton[19] reported Cs^{127} to be a positron emitter with a half life of 5.5 hours. The mass assignment was made by the identification of the 34-day Xe^{127} daughter. More recently Wapstra, Verster and

Boelhouwer[20] have studied the gamma rays of Cs^{127} by scintillation spectrometric methods and have reported gamma rays of 410 ± 20 and 125 ± 5 kev.[20]

In our experiments we noted that the cesium fraction of a calcium iodide target bombarded with 60-Mev helium ions in the 184-inch cyclotron was a mixture of 30-minute Cs^{130}, 6.25-hour Cs^{127} and 31-hour Cs^{129}. Even immediately after the bombardment, the major activity was Cs^{127} and if the isolation of cesium were delayed a few hours past the end of the bombardment more than 90 percent of the GM activity was Cs^{127}.

The mass assignment was checked by a run in the time-of-flight mass spectrometer.[21] This was done with the help of Dr. Maynard C. Michel who used the instrument described in the section on instruments. Decay curves taken with a GM counter using mass-separated Cs^{127} showed no deviation from a half life of 6.25 ± 0.1 hours in more than 4 half lives. This half life is somewhat longer than that given by Fink, et al.[19] Negligible 6.25-hour activity was collected at neighboring mass positions.

The positrons of Cs^{127} were studied in the beta-ray spectrometers briefly described in a later section. The runs were not made with mass-separated activity because sources of sufficient intensity could not be prepared. The presence of a small amount of Cs^{129}, however, caused no difficulty in the interpretation of data on unseparated cesium fractions isolated a few hours after bombardment. Our best runs were obtained on a lens spectrometer. Figure 1 shows the Fermi-Kurie plot of the positron spectrum. The most energetic component has an energy of 1.063 ± 0.10 Mev. The presence of numerous conversion electrons complicates the resolution of the second component and the value of 685 kev (378 kev lower in energy than the first component) is uncertain to perhaps 25 kev. This leaves

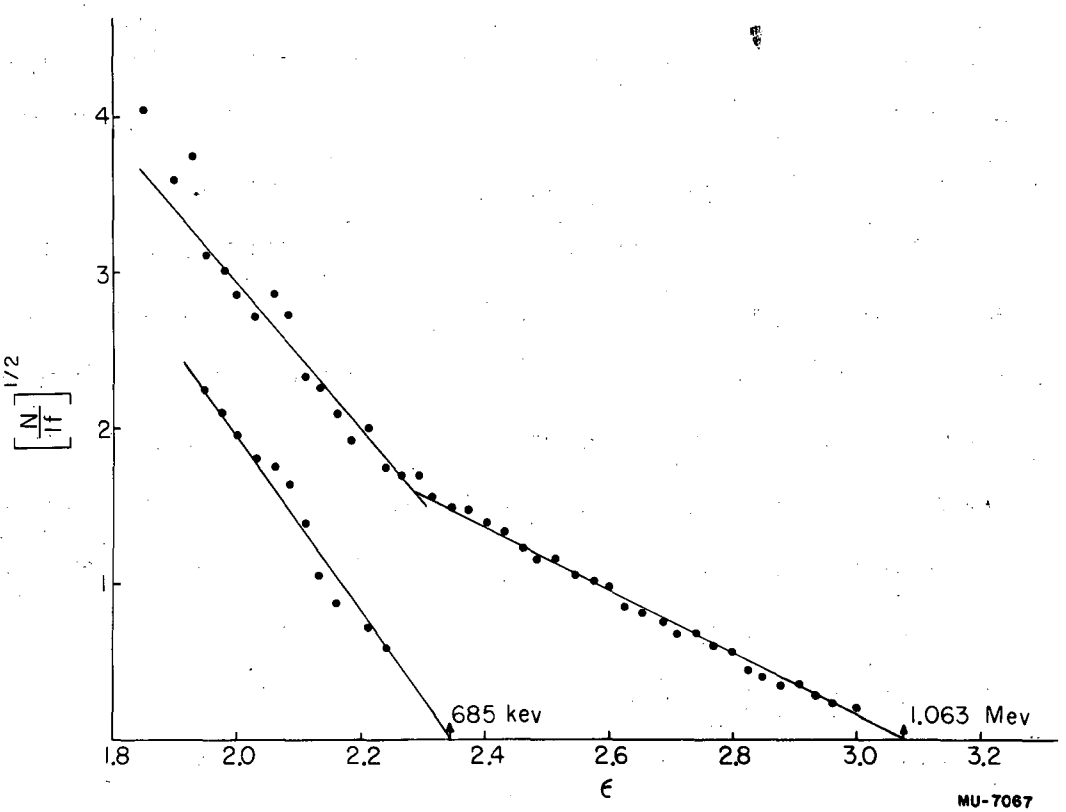

Fig. 1. Fermi-Kurie plot of high energy portion of Cs^{127} positron spectrum. Resolution of a second component after correction for conversion electrons is shown.

unresolved the question whether the prominent 406-kev gamma ray observed in the scintillation spectrometer (see below) connects the Xe^{127} energy levels reached by these two positron groups.

Figures 2 and 3 show the gamma spectrum of Cs^{127} as determined on a scintillation spectrometer. There are prominent gamma rays of energies of 125 and 406 kev as well as a large 30-kev peak of xenon x-radiation from the electron capture decay of Cs^{127}. The small amount of 510-kev annihilation radiation by comparison with this peak indicates a low β^+/EC ratio ($<1/15$). The ratio of the 406-to 125-kev gamma radiation after correction for counting efficiency is about 8.

Figures 5 and 4 show the conversion electron spectrum of Cs^{127} as studied on the magnetic double-focusing spectrometer and the lens spectrometer, respectively. In Table 1 the energies of the electrons, the conversion shell and the energy of the corresponding gamma rays are given. The K and L lines of the 125- and 406-kev gamma rays are identified. Conversion electrons of 3 or more gamma rays of lower intensity are present but the assignment of K and L lines is uncertain. Nothing further was learned about this group since their intensity was so low that no further data was obtained in the scintillation spectrometer or the coincidence spectrometer.

The gamma-gamma coincidence studies on Cs^{127} gave the following results. The 125-kev gamma radiation is not in coincidence with the 406-kev gamma radiation. It is in coincidence with 30-kev x-radiation and with 440-kev gamma radiation. The intensity of the latter was low enough to explain why it was not seen in the straight gamma runs. The 406-kev radiation was not in coincidence with 125-kev radiation, but was

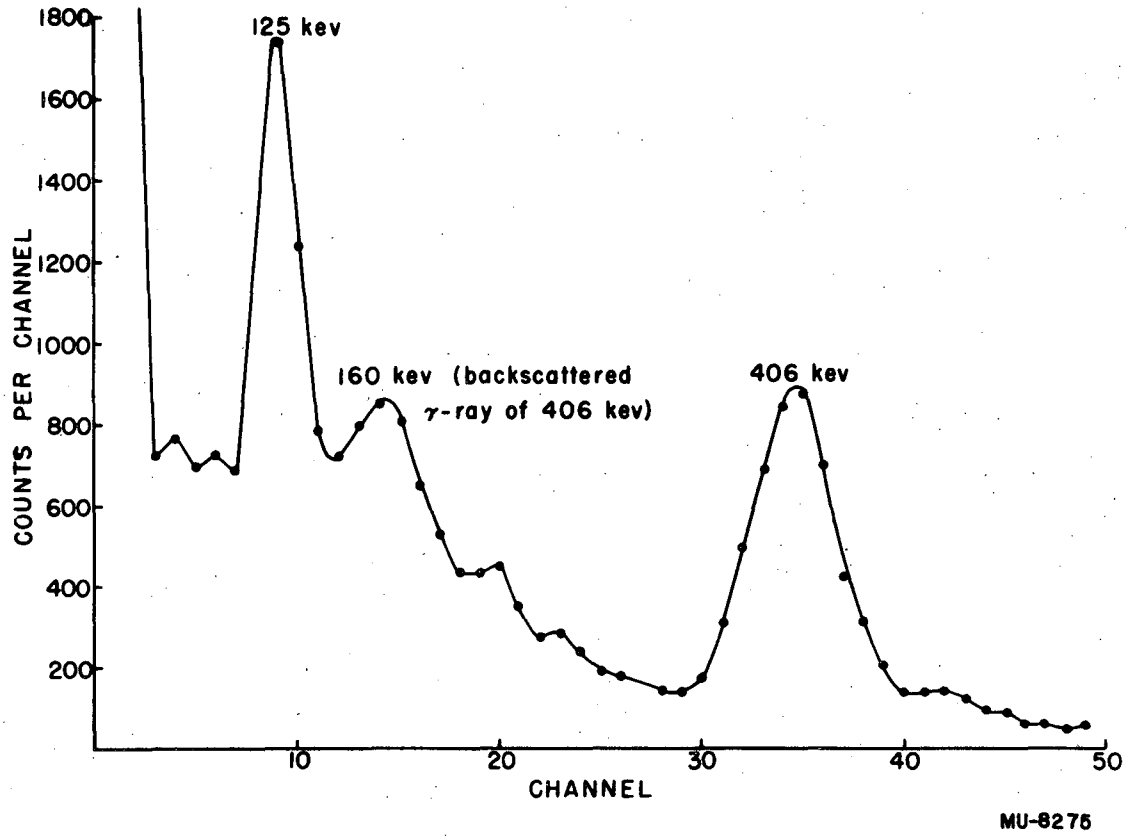

Fig. 2. Gamma spectrum of Cs^{127} determined in scintillation spectrometer. Sample mounted on aluminum.

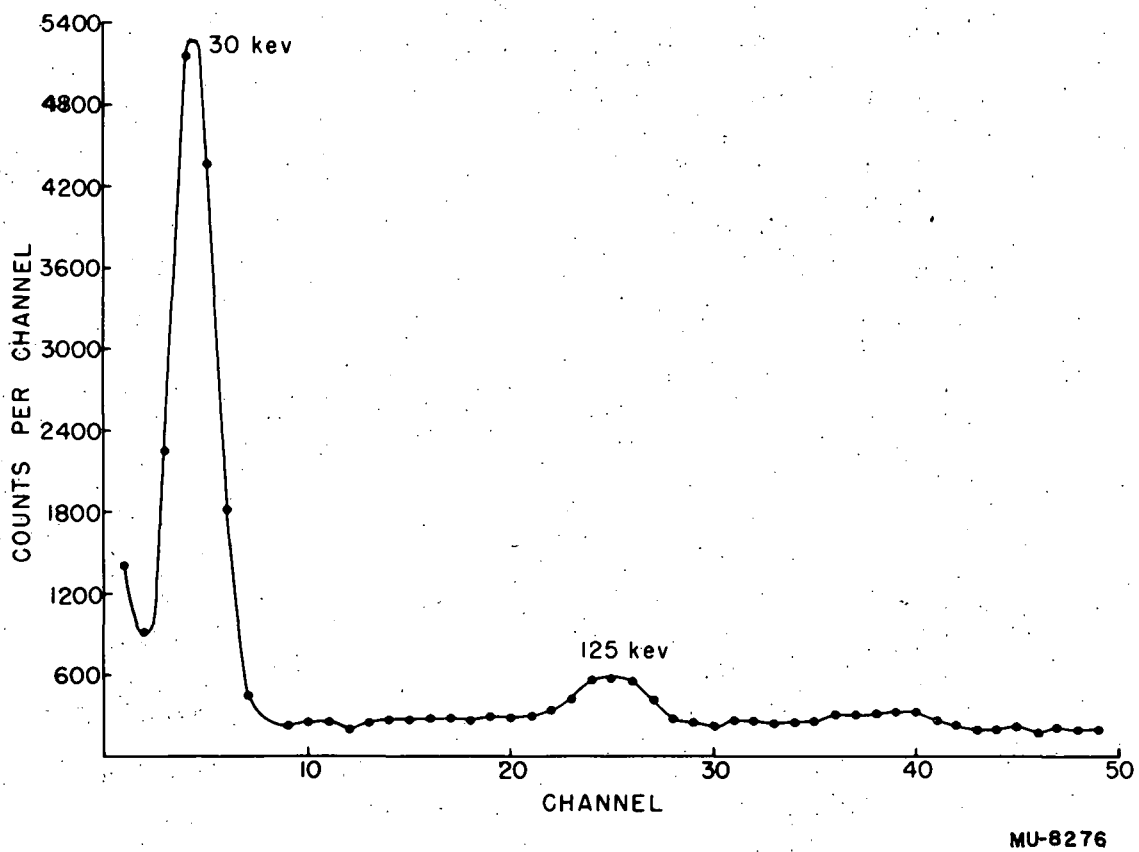

Fig. 3. Gamma spectrum of Cs^{127} in 0- to 200-kev region showing prominent x-ray peak. Sample mounted on aluminum.

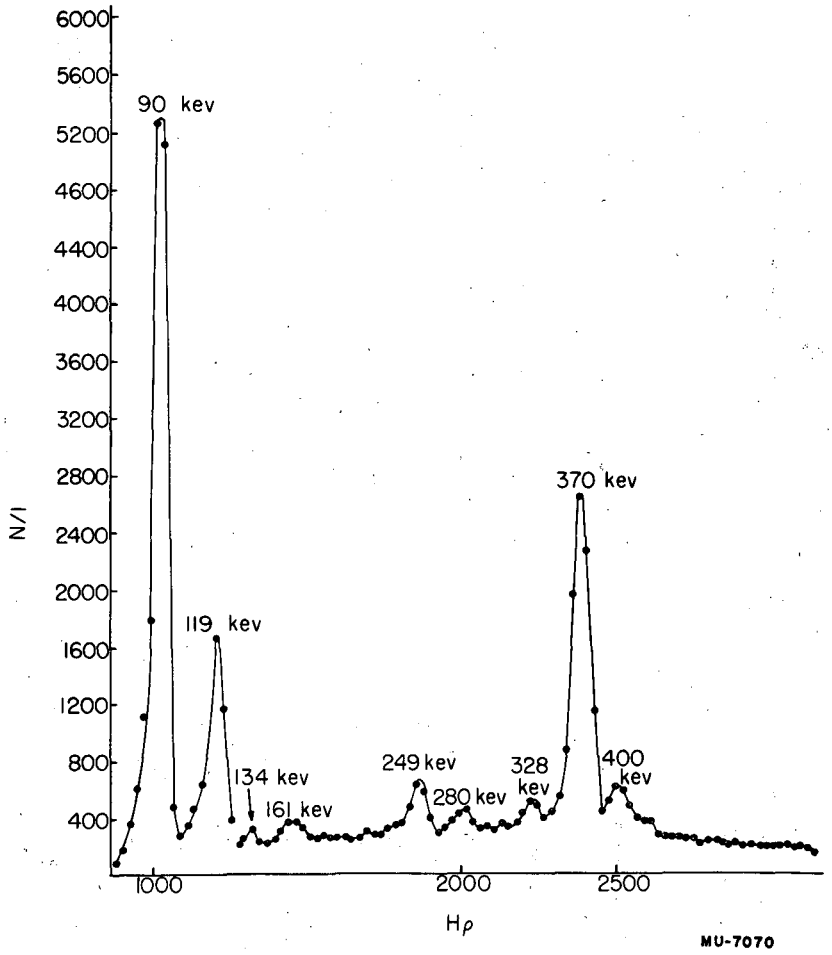

Fig. 4. Conversion electron spectrum of Cs^{127} in 80- to 450-kev region determined in lens spectrometer.

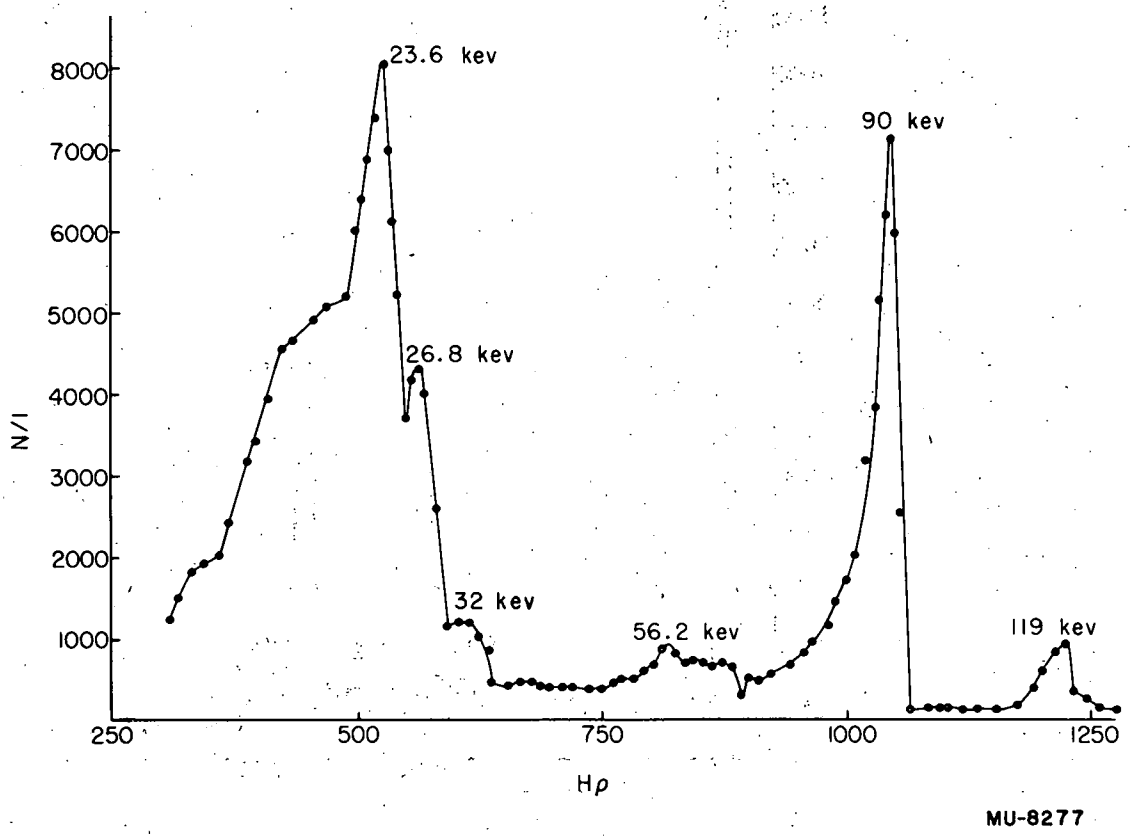

Fig. 5. Conversion electron spectrum of Cs^{127} in 0- to 120-kev region in double-focusing spectrometer.

Table 1

Conversion Electrons of Cs^{127}

Energy of electrons (kev)	Identification of conversion shell	Gamma ray energy (kev)	K/L+M ratio
23.6	Auger electrons (KLL)	-	-
26.8	Auger electrons (KLY)	-	-
32.0	Auger electrons (KXY)	-	-
56.8	K conversion (?)	-	-
90.0	K conversion	125	7.9
119	L conversion		
134	K conversion (?)	169 (?)	-
161	K conversion (?)	196 (?)	-
249	K conversion (?)	285 (?)	-
280	L conversion (?)		
328	K conversion (?)	363 (?)	-
370	K conversion	406	6.3
400	L conversion		

in coincidence with 30-kev x-radiation. In coincidence experiments in which annihilation radiation was selected by the gate crystal, coincidence peaks were observed at 125 and 406 kev.

These findings in connection with the information derived from the isomer studies to be described next were employed in the construction of the proposed decay scheme shown in Fig. 10 (see page 31). Following Wapstra[22] we designate the ground state of Cs^{127} as $d_{5/2}$. We have included a 280-kev gamma ray in Fig. 10 to connect the $g_{7/2}$ and $d_{5/2}$ levels. The electron lines at 249 and 280 kev (see Table 1 and Fig. 5) may be the conversion electrons of this transition but the gamma intensity was too low for resolution from the scintillation spectrometer curves. In the coincidence studies the 125-to 280-kev coincidence to be expected from Fig. 10 was looked for but unfortunately it was completely obscured by a large spurious 125- to 280-kev coincidence resulting from Compton scattering of the 406-kev gamma ray.

A number of preliminary experiments made it evident that the decay of Cs^{127} to the 75-second isomeric state of Xe^{127} was slight if it took place at all. This isomer had been reported by Creutz et al.[23] who had bombarded iodide targets with 5- to 6-Mev protons to make Xe^{127} by the (p,n) reaction. In the volatile fraction removed from the target these workers had found two activities of 34 ± 2 days and 75 ± 1 seconds, respectively, and assigned both to Xe^{127}.

Studies of the conversion electrons emitted by the 75-second isomer had shown 3 groups of 91.4, 140 and 170 kev, respectively. (A permanent magnet spectrometer with photographic emulsion detection was used.) The first was interpreted as the K line of a 125-kev gamma ray and the last

two as the K and L lines, respectively, of a 175-kev gamma ray. Secondary x-radiation resulting from the conversion of these gamma rays was also observed.

We were able to observe this isomer in the decay of Cs^{127} in the following manner. An active sample of carrier-free Cs^{127} was evaporated on a metal disk and placed in the depression of the slide of the apparatus diagrammed in Fig. 6. The greased tight-fitting slide was pushed in until the cesium sample was located in the 1.5-inch diameter chamber 0.25-inch deep, drilled in from the top of the block. This chamber was covered with a 0.1-mil foil of aluminum to prevent loss of xenon. After a growth period of 3 minutes the Cs^{127} sample was withdrawn. Recoil daughter atoms of Xe^{127} and Xe^{127m} ejected from the cesium source plate during the growth period remained in the closed chamber and their radiations were examined with a sodium iodide crystal mounted immediately above the chamber. The whole assembly was housed in a 2-inch thick lead castle. The gamma spectrum was determined with a 50-channel analyzer making runs at about 1 minute intervals for 10 or 15 minutes.

Figure 7 shows the spectrum of the recoil activity obtained immediately after the growth period. Two gamma rays of 125- and 175-kev energy are to be noted as well as xenon K x-rays. Since the decay of all 3 peaks followed a 75-second decay line (see Fig. 8) there is little doubt that this is the same activity as that observed by Creutz *et al.*[23] It was established more securely that this isomer was the product of the decay of Cs^{127} by plotting the yield of the isomer in a 3-minute growth period as a function of time at which the "milking" took place. The half life of 6.1 hours so obtained (as shown in Fig. 9) confirms the genetic relationship.

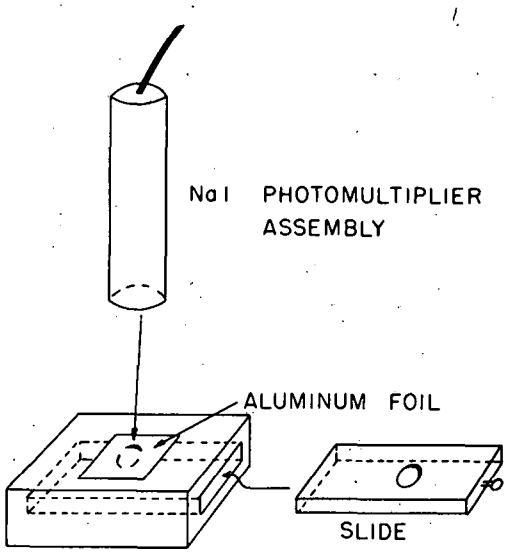

Fig. 6. Apparatus for the collection and study of the gamma spectrum of short-lived xenon daughter activity from Cs^{127} or Cs^{125} samples.

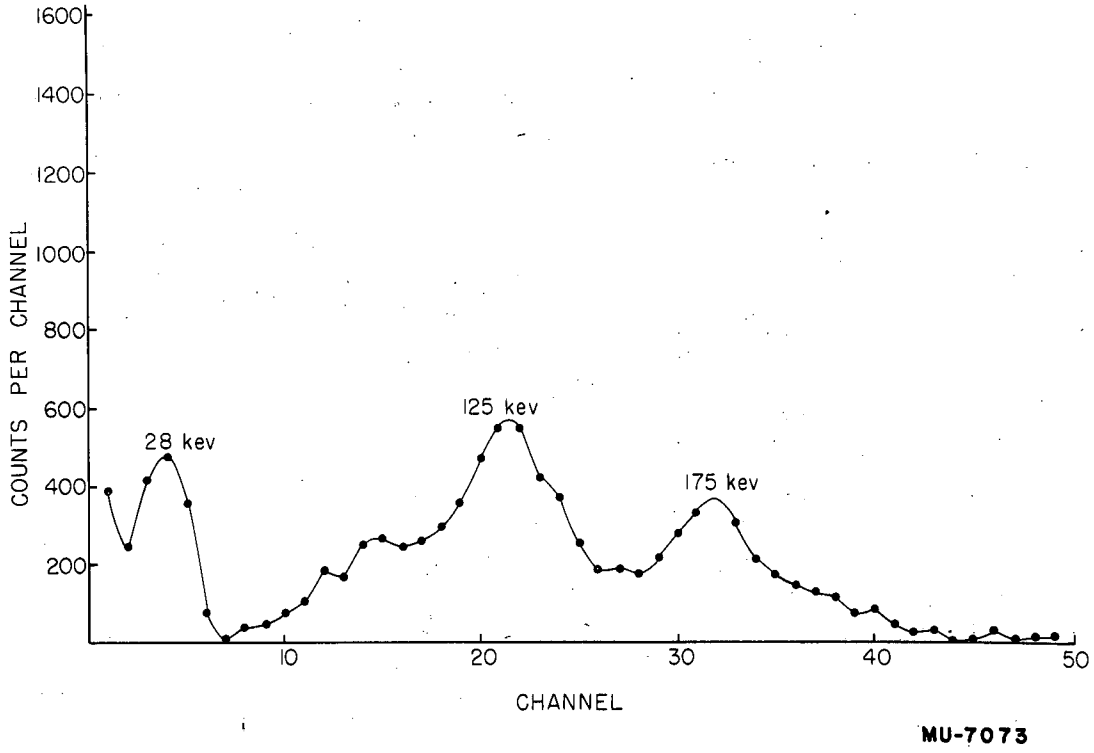

Fig. 7. Gamma spectrum of Xe^{127m}.

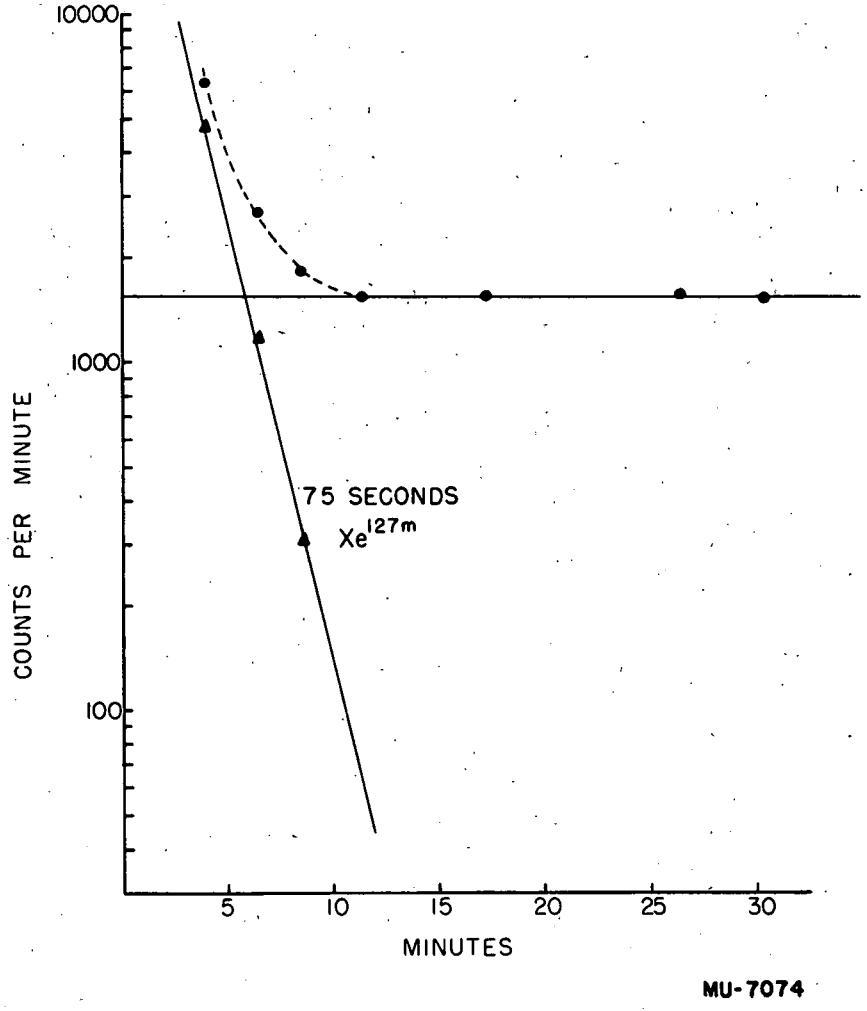

Fig. 8. Decay of activity in gamma peaks showing resolution of 75-second activity.

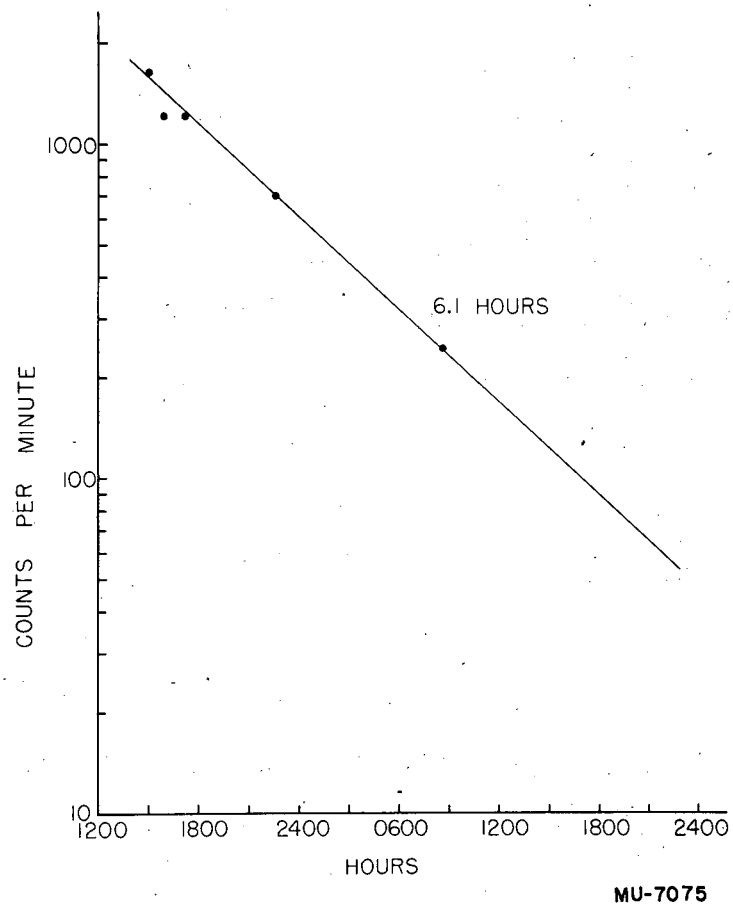

Fig. 9. Yield of 75-second Xe^{127m} from timed milkings of Cs^{127} parent sample.

A small amount of long-lived recoil activity remains after decay of the 75-second Xe^{127m}. If all of this is assigned to 34-day Xe^{127}, the branching of Cs^{127} to 34-day Xe^{127} and to 75-second Xe^{127m} is estimated to be in the ratio of $\sim 10^4/1$.

In their summary of nuclear isomers Goldhaber and Sunyar[4] listed Xe^{127m} as an E3 isomer with 175-kev gamma ray indicated as a $h_{11/2}$-$d_{5/2}$ transition. They preferred to reinterpret tentatively the 91-kev electrons observed originally by Creutz et al.[23] as L electrons of a 96-kev $d_{5/2}$-$s_{1/2}$ E2 transition, rather than as K electrons of a 125-kev transition. Their reason for this reassignment was the nonobservation of L electrons of the 125-kev gamma ray which should have prominent K/L ratio (~1 to 2) for a 125-kev E2 transition. More recently Wapstra, Verster and Boelhouwer[20] in discussing the 125-kev radiation prominent in the decay of Cs^{127} stated that it was probably identical with the second transition in the decay of 75-second Xe^{127m} and that the 125-kev gamma ray represents an M1 transition going to the $d_{3/2}$ ground state in Xe^{127}. Our result seems to confirm this interpretation. The K/L+M ratio of 7.9 obtained by us for the 125-kev gamma ray in the decay of Cs^{127} is of the correct magnitude for M1 and much too high for an E2 radiation.

The Xe^{127m} results have been incorporated in the decay scheme of Fig. 10.

The longer-lived Xe^{127} formed in the decay of Cs^{127} is the 34-day xenon activity studied by Bergström[18] and several other workers. To get larger samples for study than were available by decay of our Cs^{127} samples, Xe^{127} was prepared by the (p,n) reaction on I^{127} in the form of

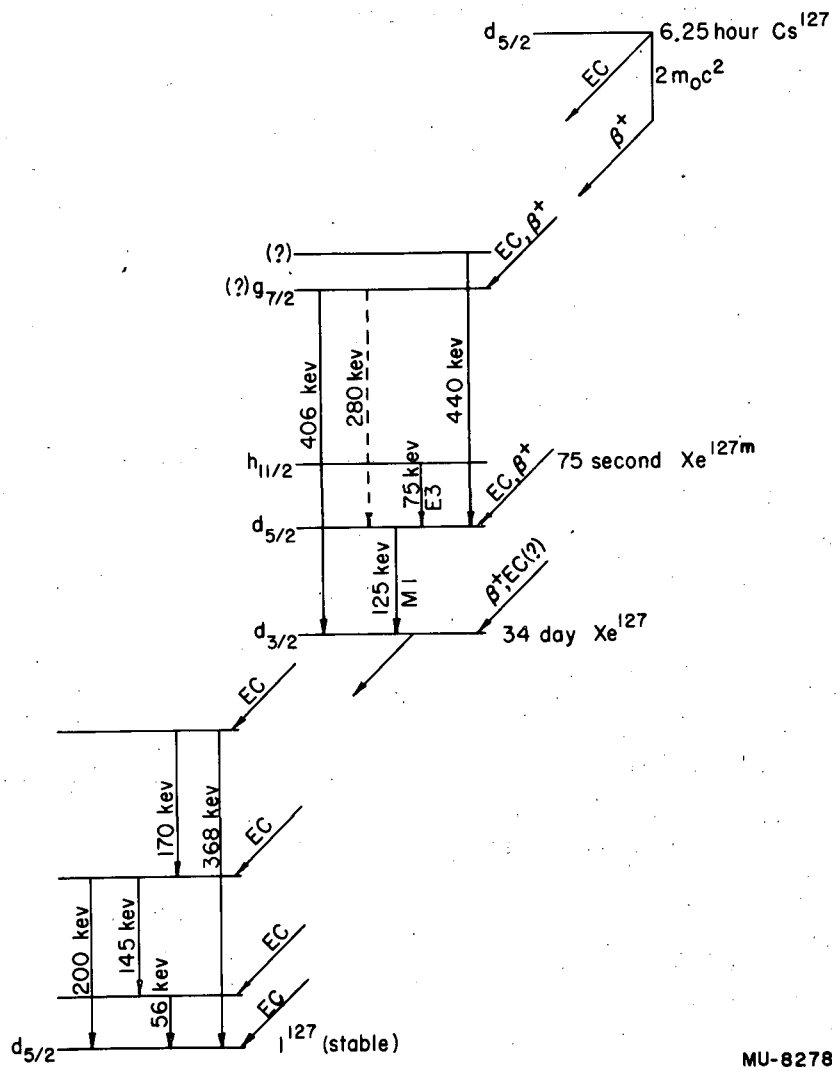

Fig. 10. Proposed decay scheme of Cs^{127}.

potassium iodide and by isolation of xenon by the glow-discharge method described in a later section.

Bergström[18] studied the electron spectrum of mass-separated Xe^{127} in detail and reported gamma rays at 56, 145, 170 and 200 kev. His scintillation spectrometric studies could not resolve the gamma rays of energies 145, 170 and 200 kev, but he found an additional gamma ray of 365 kev, the conversion electrons of which were not detectable with the beta spectrometer.

The scintillation spectrometer studies on Xe^{127} were repeated. Figure 11 shows the gamma spectrum of Xe^{127} obtained by a sodium iodide crystal spectrometer coupled to a 50-channel differential-pulse analyzer. Photopeaks at 56, 170, 200 and 368 kev were observed in agreement with Bergström. The small peak at 75 kev is due to lead x-rays produced by the lead castle enclosing the sodium iodide-photomultiplier assembly, while the peak at 110 kev is due to backscattered radiation of the most prominent gamma peak, at 200 kev, in the gamma spectrum of Xe^{127}. The gamma ray of 145 kev was not detectable in the gamma spectrum because of its low intensity, but in our gamma-gamma coincidence studies (see below), a prominent peak at 145 kev in addition to that at 170 kev was obtained in the coincidence spectrum, when the gamma ray selected by the gate was 56 kev (see Fig. 12).

The gamma-gamma coincidence studies established that the 56-kev gamma ray was in coincidence with 145- and 170-kev gamma rays, and with 28-kev iodine x-rays, as suggested by Bergström by the fact that the sum of the energies of 145 and 56 kev is 201 kev. In agreement with this, the 200-kev radiation is not in coincidence with 56- and 145-kev gamma

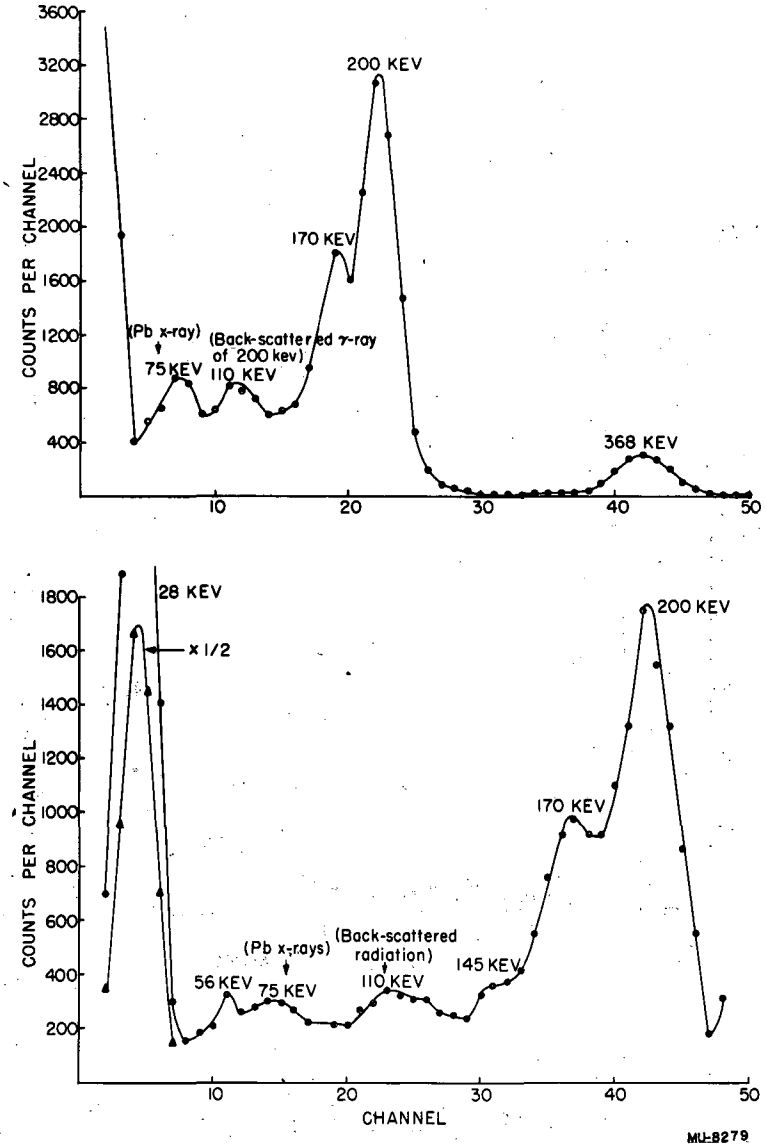

Fig. 11. Gamma spectrum of 34-day Xe^{127}.

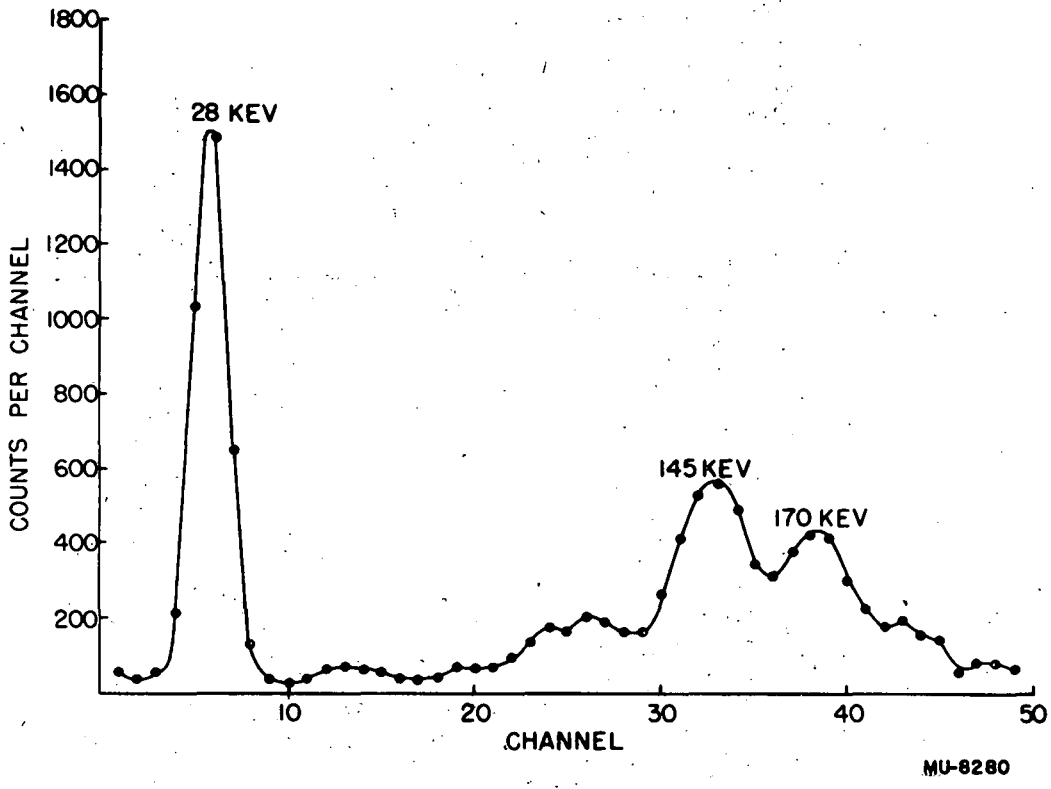

Fig. 12. Gamma spectrum of Xe^{127} showing the gamma rays in coincidence with the 56-kev gamma ray.

radiations, but is in coincidence with the 170-kev gamma ray and the 28-kev iodine x-radiation. Quantitative studies indicate that 56-kev radiation should be below 145-kev radiation, while 170-kev radiation should lie higher than 200-kev radiation. For example, in the study of 170- to 200-kev coincidence the ratio of coincidence pulses to gate pulses was greatly increased when the gamma ray selected by the gate crystal is changed from 200 to 170 kev. The 368-kev gamma ray is in coincidence with only 28-kev iodine x-radiation and with no other gamma rays. Because most of the coincidence measurements were straightforward the curves are not presented. In Fig. 12, however, the gamma spectrum in coincidence with the 56-kev gamma ray is shown because it indicates that the 145-kev gamma ray is clearly evident. This gamma ray was not resolved in the gamma curves of Fig. 11. All coincidence results are incorporated in the decay scheme of Fig. 10.

B. The Radiations of Cs^{125} and the Isomer Xe^{125m}

In the bombardments of calcium iodide with helium ions a new positron activity of 45-minute half life appeared in the cesium fraction when the energy of the bombarding helium ions was raised to 100 Mev. One hour after the end of bombardment this new activity accounted for more than 80 percent of the GM activity in the cesium fraction, most of the rest being 6.25-hour Cs^{127} and a small amount of 31-hour Cs^{129}. When the xenon daughter activity was separated and deposited by the glow-discharge method on metallic counting foils, a GM counter showed the presence of a mixture of 18-hour Xe^{125} and 34-day Xe^{127}. This suggested that the 45-minute activity was Cs^{125} and the assignment was confirmed by mass

separation. A GM-decay curve of the mass 125 fraction isolated with the help of Dr. M. C. Michel in the time-of-flight mass separator showed a straight line decay of 45 ± 1 minutes from an initial counting rate of 18,000 to less than 100 counts per minute. The counting efficiency of the 18-hour electron capture daughter Xe^{125} was very low in the GM tube but when the sample was placed in a "nucleometer" windowless methane-flow proportional counter a decay line of 18-hour half life was observed.

It was possible to determine the energy of the most energetic positron group of Cs^{125} by running a cesium sample in the double-focusing beta-ray spectrometer starting about 1 hour after the bombardment because this positron energy was greater than that of Cs^{127}. Figure 13, which shows a Fermi-Kurie plot of the high energy region, indicates that the energy of the Cs^{125} positrons is 2.05 ± 0.02 Mev. Because of the presence of Cs^{127} no attempt was made to resolve out lower energy components.

The gamma spectrum of Cs^{125} (see Figs. 14 and 15) shows a gamma ray of 112 kev in addition to annihilation radiation and 30-kev xenon x-rays. No gamma rays of higher energy were observed. The large abundance of x-rays as compared with the gamma rays suggests that Cs^{125} decays prominently by electron capture but the electron capture/positron branching ratio was not determined. As in the case of Cs^{127}, gamma-gamma coincidence studies were carried out on Cs^{125}. It was observed that the 112-kev gamma ray is in coincidence with K-radiation and also with annihilation radiation.

The conversion electron spectrum was not studied in detail but the energy of the 112-kev gamma ray was checked by measurement of the K and L conversion lines. The K/L ratio was determined as 3.6. This may be

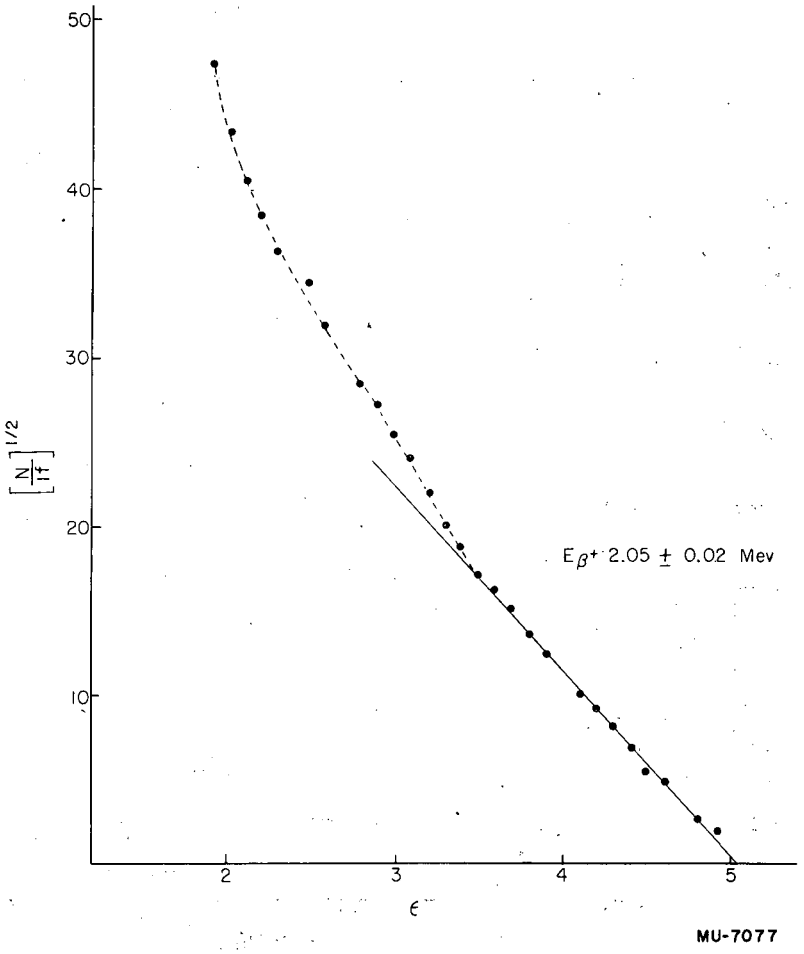

Fig. 13. Fermi-Kurie plot showing endpoint energy of the most energetic positrons in Cs^{125}.

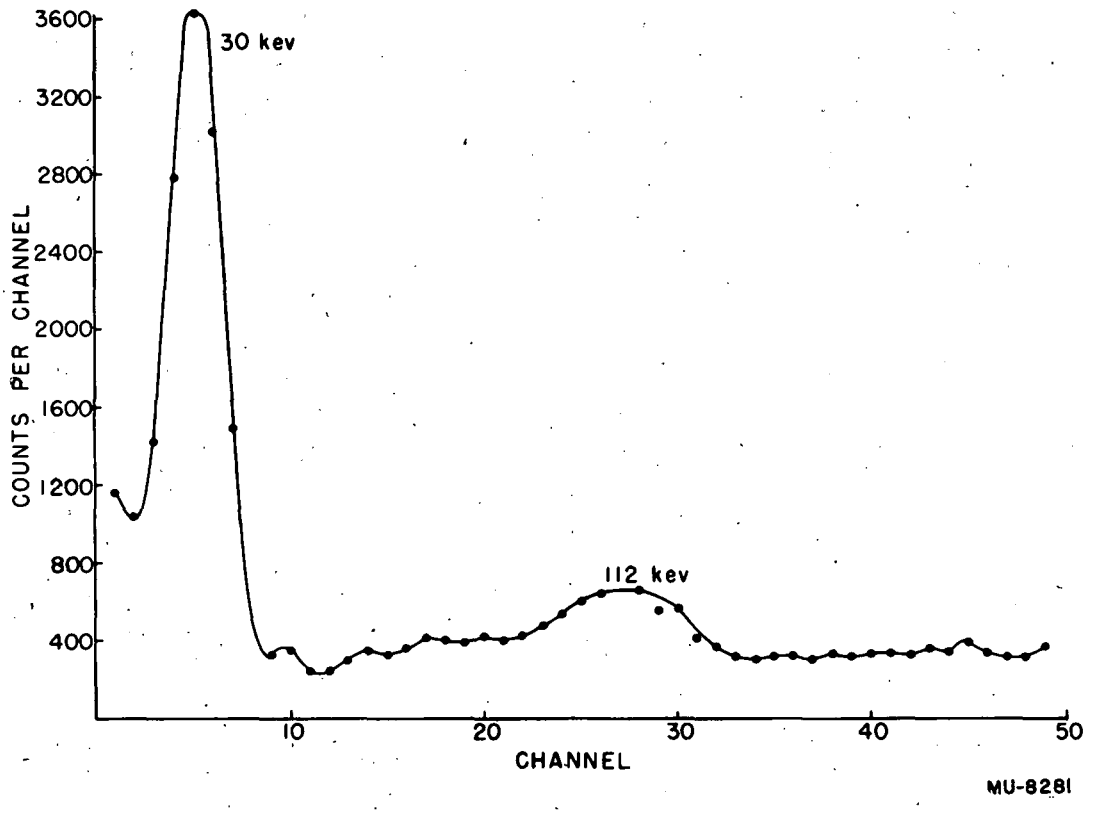

Fig. 14. Gamma spectrum of Cs^{125} in 0- to 200-kev region. Sample mounted on aluminum.

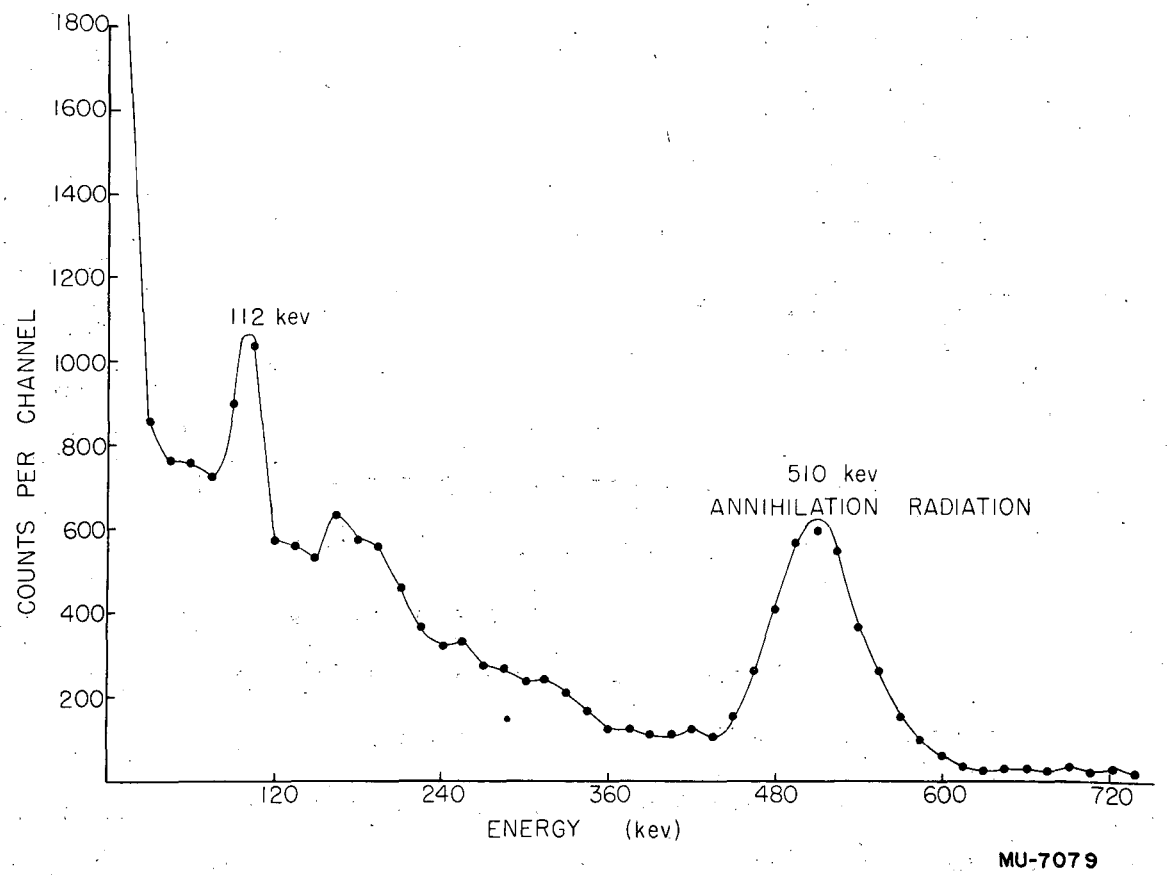

Fig. 15. Gamma spectrum of mass-separated Cs^{125} in 0- to 600-kev region.

somewhat in error since the time spent in obtaining the data made necessary rather large half-life corrections.

It is believed that the 112-kev transition is the $d_{5/2}$-$d_{3/2}$ transition in Xe^{125} analogous to the 125-kev transition in Xe^{127}. From Goldhaber and Sunyar's[4] empirical curves one would expect a K/L ratio of 7 to 8 for an M1 transition of this energy.

At the suggestion of Dr. Ingmar Bergström we made a search for an isomer of Xe^{125} because his study of the systematics of the odd-mass xenon isotope,[18] as well as those of Goldhaber and Hill,[5] indicated the probability of a short-lived E3 isomer in Xe^{125} very similar to that in Xe^{127}. By performing experiments analogous to those described above in the Xe^{127m} case we have been successful in the search for this isomer, but because the interference from 18-hour Xe^{125} was much greater than that from 34-day Xe^{127} in the earlier study, the results are not quite as clean-cut.

Carrier-free cesium samples isolated within 1 hour of the end of bombardment from calcium iodide targets bombarded with 100-Mev protons were inserted in the depression of the slide of Fig. 6 and pushed into the inner recoil collection chamber for 1-minute growth periods. Immediately after the cesium source was removed the gamma spectrum of the xenon daughter recoil activity was studied with the sodium iodide 50-channel analyzer. The registers were photographed with a Leica camera at 12-second intervals over a period of several minutes without disturbing the count switch. By subtraction of the dial readings of one negative from that of the next the spectrum during any particular 12-second period could be reconstructed. The upper curve of Fig. 16 shows the spectrum integrated

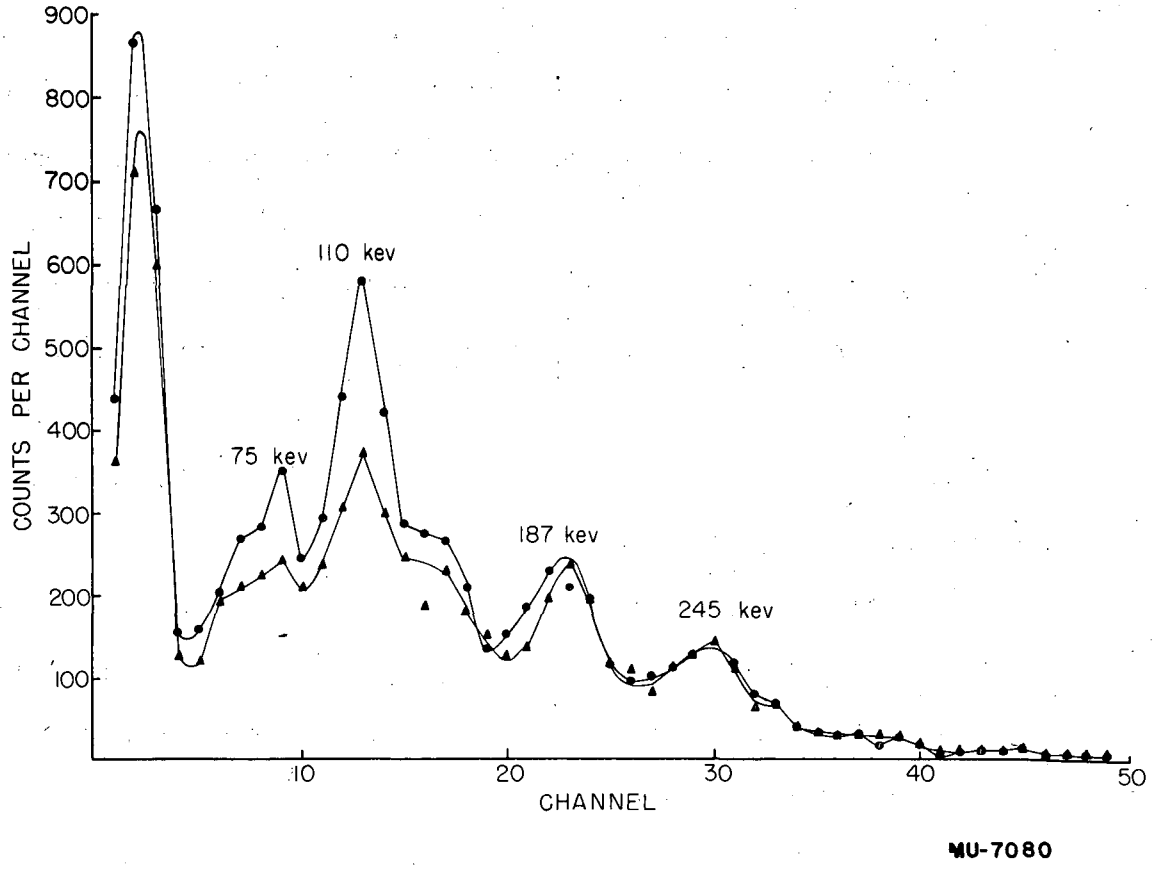

Fig. 16. Gamma spectrum of Xe^{125m} + Xe^{125} daughter activity collected in apparatus of Fig. 7 during 1-minute growth period. Upper curve is the spectrum recorded for first one minute after collection; lower curve is for second minute.

over the first 1 minute, while the lower curve shows the spectrum of the second minute's run. It is seen that gamma rays of 75, 110, 187 and 245 kev as well as 30-kev x-radiation are present.

As shown in Fig. 17 the areas under the x-ray peak, the 75-kev peak and the 110-kev peak decay intially with a half life of 50 to 60 seconds and this decay in all likelihood is correctly assigned to Xe^{125m}. (The longer-lived component in the curves of Fig. 17 is based on points not shown.) Part of the radiation at these energies and most or all of it at 187 and 245 kev is due to 18-hour Xe^{125} as will be discussed.

We are inclined to the view that the 110-kev peak is identical with the 112-kev peak of Figs. 14 and 15 and that it represents the M1 transition, $d_{5/2}$-$d_{3/2}$, following a 55-second E3 transition, $h_{11/2}$-$d_{5/2}$. The 75-kev radiation may be this E3 radiation as indicated in Fig. 18, but we cannot entirely exclude the possibility that this is fluorescence radiation from the lead shielding and that the true E3 radiation is hidden in the region above 110 kev.

One difficulty in the further study of the isomer is the low-branching ratio for its production in the decay of Cs^{125}. This branching ratio was estimated by comparing the amount of 187-kev gamma radiation of Xe^{125} with the amount of 110-kev gamma radiation of Xe^{125m} with suitable corrections for half life and for the length of the growth period. Making the assumption of one 187-kev gamma ray per decay of Xe^{125} the branching decay of Cs^{125} to Xe^{125m} is $\sim 10^{-3}$.

A good part of the information on the decay of Cs^{125} and Xe^{125m} is summarized in the proposed decay scheme of Fig. 18. It can be observed that the decay schemes of Cs^{125} and Cs^{127} are very similar.

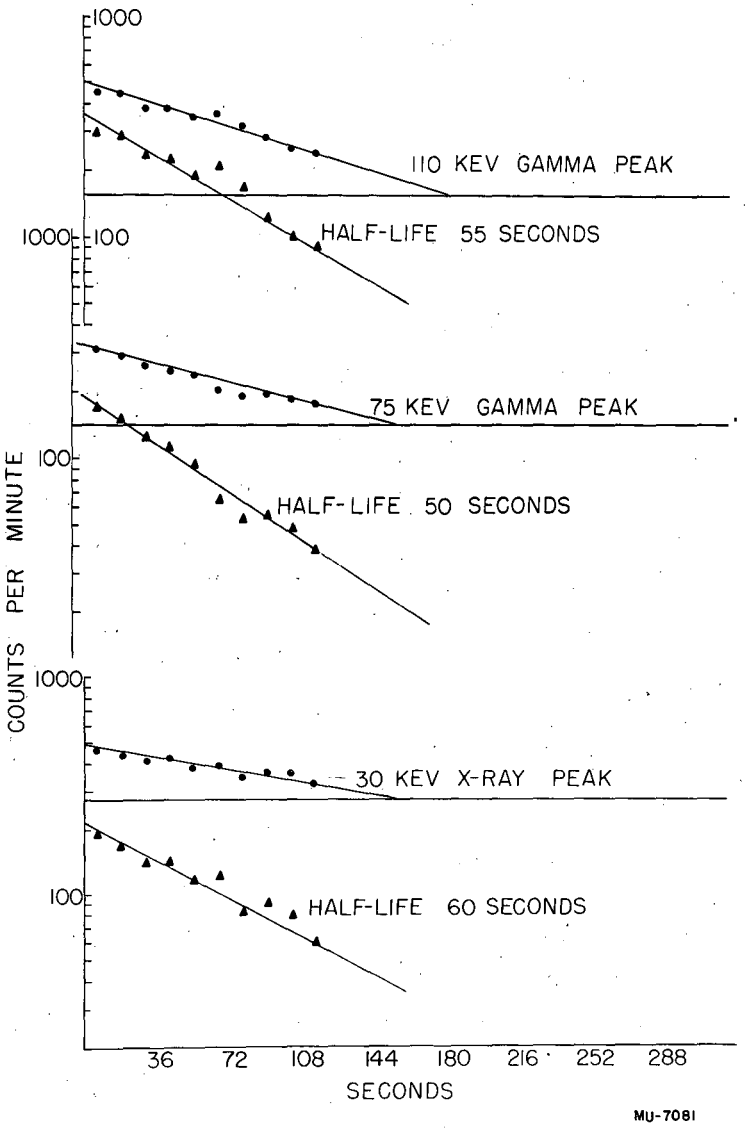

Fig. 17. Decay of gamma rays in 30-, 75- and 110-kev peaks during 12-second run on Xe^{125m} + Xe^{125} daughter activity collected from Cs^{125} source. Resolution shows 55-second decay of Xe^{125m}.

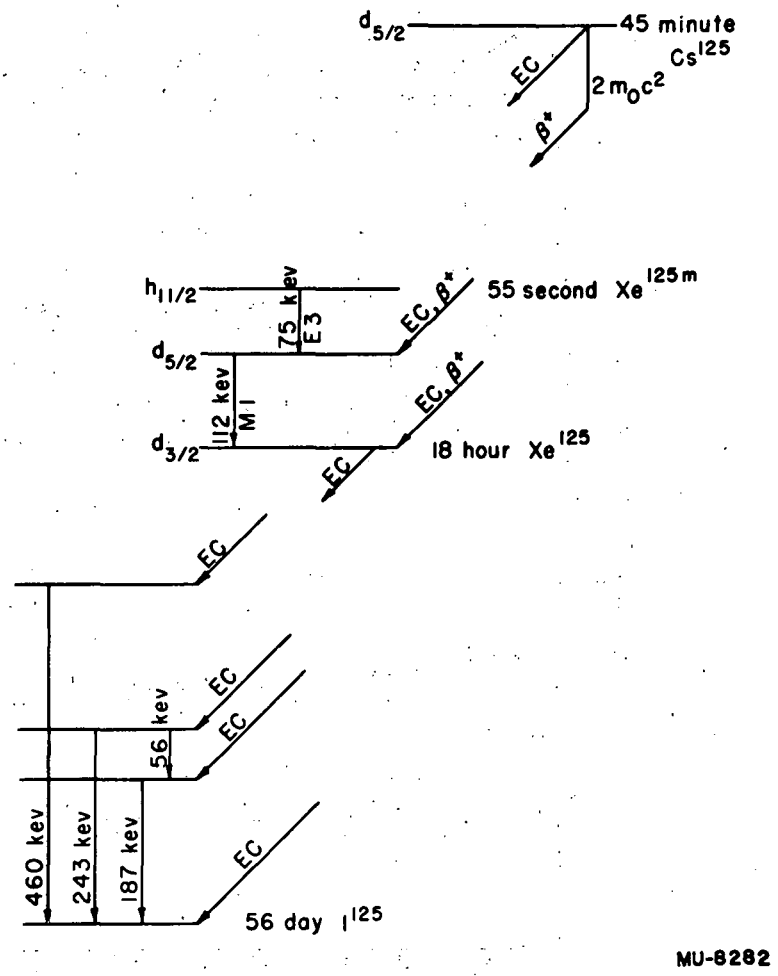

Fig. 18. Proposed decay scheme for Cs^{125}.

The longer-lived Xe^{125} formed in the decay of Cs^{125} is identical with the 18-hour activity mass-separated and characterized by Bergström.[18,24] We repeated Bergström's scintillation spectrometer studies (but not his conversion electron studies) using Xe^{125} samples isolated by the glow-discharge method from decayed Cs^{125} samples.

Figure 19 shows the gamma spectrum determined in the scintillation spectrometer. Definite photopeaks were observed at 56, 187 and 243 kev in agreement with Bergström. The small peak at 110 kev is probably Compton radiation from the 187-kev gamma ray. No annihilation radiation was observed. In the coincidence spectrometer it was established that the 56- and 187-kev gamma rays are in coincidence as was suggested by Bergström by the fact that the energies sum to 243 kev. In agreement with this the 243-kev gamma ray was not in coincidence with the 56- or 187-kev gamma rays. The 56-kev gamma ray must lie higher than the 187-kev gamma ray because the ratio of coincidence pulses to gate pulses increased greatly when the gamma ray selected by the gate was changed from 187 to 56 kev. The 96- and 106-kev gamma rays whose conversion electrons were observed by Bergström were too low in intensity to observe in our coincidence studies. We can set an upper limit of 2 percent for these gamma rays compared with the 187- and 243-kev gamma rays when the latter are applied to the gating circuit. These results have been incorporated in the decay scheme shown in Fig. 18.

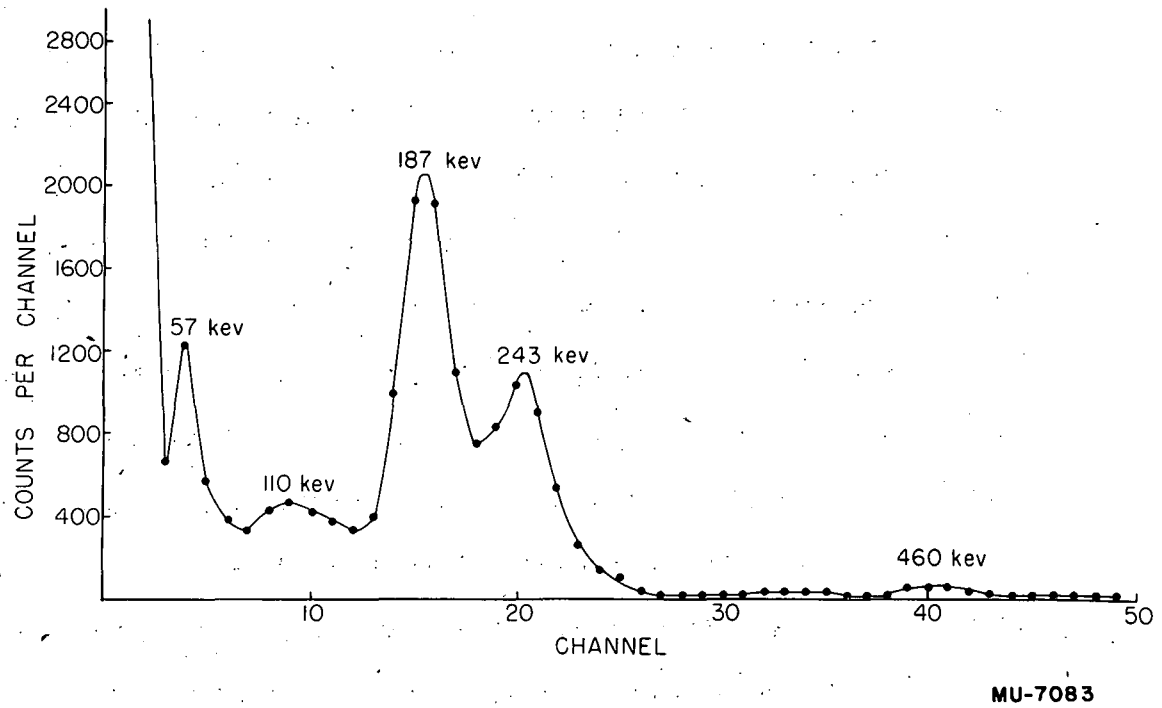

Fig. 19: Gamma spectrum of 18-hour Xe^{125}.

C. Cesium 123

In calcium iodide targets bombarded with helium ions of 130 Mev a new cesium activity of 6-minute half life was produced along with 45-minute Cs^{125}. A small amount of 6.25-hour Cs^{127} is also produced. When the xenon daughter activity grown in during a 5-minute period was isolated and deposited on thin metallic counting foils by the glow-discharge method and followed for decay in a GM counter, a mixture of 1.8-hour Xe^{123} and 18-hour Xe^{125} was observed. When this procedure was repeated after complete decay of the 6-minute cesium parent, only 18-hour Xe^{125} was isolated and no 1.8-hour Xe^{123} activity was observed in the decay of the isolated xenon daughter sample. This identifies the 6-minute cesium activity as Cs^{123}. Annihilation radiation of 6-minute half life was prominent in scintillation spectrometer curves taken on the cesium fraction shortly after the bombardment indicating that Cs^{123} decays at least in part by positron emission. Because of the short half life of Cs^{123}, no detailed study was carried out.

D. Cesium 130

Following the preliminary studies of Risser and Smith,[25] and of Fink, Reynolds and Templeton[19] on a 30-minute cesium activity produced in bombardments of iodine with low-energy helium ions, a careful study of the radiations emitted by this activity was carried out by Smith, Mitchell and Caird.[26] There is little doubt that this activity is produced by an (α,n) reaction and hence the mass assignment is correctly assigned to mass number 130. Nevertheless, it is probably worth recording the confirmation of this assignment by the use of a mass spectrograph.

Iodine in the form of calcium iodide was bombarded with 20-Mev helium ions in the 60-inch cyclotron. The cesium fraction was isolated in a carrier-free form within 1 hour of the end of the bombardment and with the help of Dr. M. C. Michel of this laboratory it was run in the time-of-flight mass spectrometer. The 30-minute activity was collected in the 130-mass position.

III. STUDIES ON THE ISOMERIC PAIR Nb^{89m} AND Nb^{89}

After the study of nuclear isomerism in xenon isotopes just discussed an attempt was made to look for isomers in the nuclei near the closed shell of 50 protons or neutrons, because the single-particle model predicts two lowest-lying levels in the states $g_{9/2}$ and $p_{1/2}$ differing in spin by 4.

Levine and Hyde[27] of this laboratory, during the course of their study of neutron-deficient isotopes of niobium produced by proton bombardment of zirconium metal with 40-Mev protons, obtained evidence for a new niobium activity of half life of 1.9 hours. The GM-decay curves on the niobium fraction isolated from the target within an hour of the end of the bombardment indicated that more than 80 percent of the activity was of 1.9-hour half life, most of the rest being 14.6-hour Nb^{90} and a small amount of longer-lived activity, principally 79-hour Zr^{89} from the decay of Nb^{89}. Measurements in a beta-ray spectrograph of low resolution showed that this 1.9-hour activity consisted of protons of about 3-Mev energy. The isolation of 79-hour Zr^{89} activity from the purified niobium fraction proved the presence of Nb^{89} parent activity. Quantitative milkings of the zirconium daughter activity showed that the Nb^{89} half life was about 2 to 4

hours and hence Nb^{89} was identified with a 1.9-hour nearly 3-Mev positron.

Per Kofstad[28] during the course of his studies on the spallation products produced by bombarding silver metal with 340-Mev protons, also isolated a niobium activity of 1.9-hour half life and 3-Mev positrons. In the present study very similar results were obtained by studying the niobium fraction isolated from niobium foils bombarded with 90-Mev protons or from yttrium oxide targets bombarded with 60- to 100-Mev helium ions. In every case the two chief activities were 1.9-hour Nb^{89} and 14.6-hour Nb^{90}. The ratio by activity of Nb^{89} to Nb^{90} was roughly the same (nearly 2/6) in each case.

A. Gamma-Ray Studies Establishing the Isomerism

Niobium 89 has 41 protons and according to the single-particle shell model is likely to exist in the states $g_{9/2}$ or $p_{1/2}$, suggesting the possible occurrence of an M4 isomer. The $g_{9/2}$-$p_{1/2}$ level separation for the odd-mass isomers of niobium in the mass region 91 to 97 is given by Fig. 75 of a review article on nuclear isomerism by Goldhaber and Hill.[5] They suggest that this separation in Nb^{89} is very slight and the decision as to which spin state is the ground state is quite uncertain. Consequently, one might expect the upper state of Nb^{89} to deactivate predominantly by positron emission rather than by gamma emission. Zirconium 89 is known to exist in two isomeric forms by virtue of its 49 neutrons which gives it an odd neutron immediately below the 50-neutron shell. The upper state is the $p_{1/2}$ state and it has a half life of 4.4 minutes. Its predominant decay is by emission of a 588-kev gamma ray to the ground state (see Fig. 20).

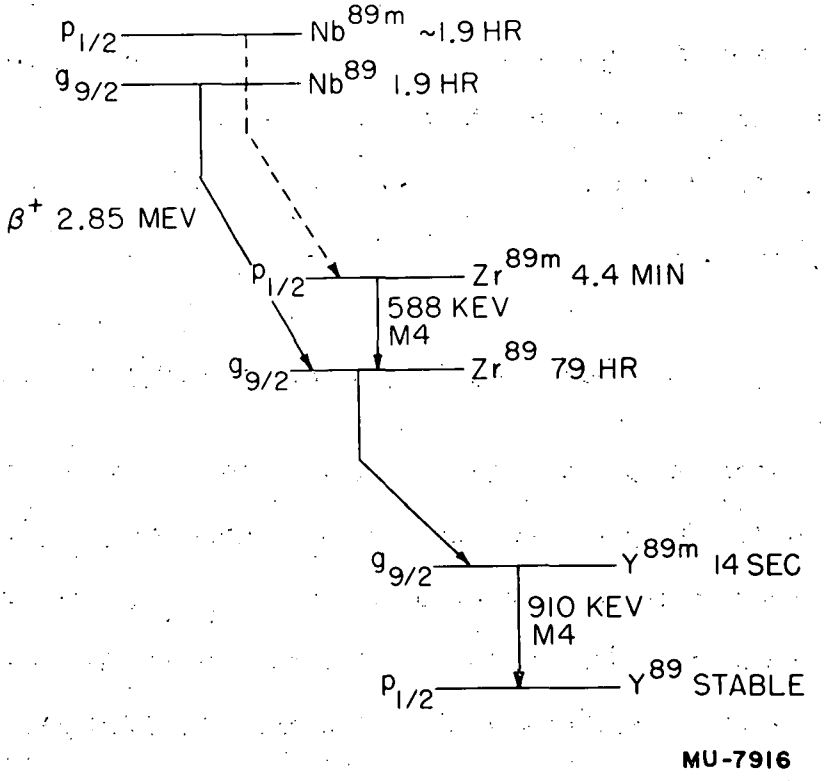

Fig. 20. Decay scheme of Nb^{89} and Nb^{89m}.

This ground state of spin $p_{1/2}$ decays with a half life of 79 hours to 13-second Y^{89m}. A pair of isomers occurs again at Y^{89} because of the odd proton below the 50-proton shell. The Y^{89m} $(g_{9/2})$ decays by emission of a 910-kev gamma ray to a stable Y^{89} $(p_{1/2})$.

Knowing these facts it was possible to design experiments to check for the expected isomerism in Nb^{89}. These experiments consisted chiefly in the examination of the zirconium daughter activity milked from niobium for the 588-kev gamma radiation of Zr^{89m} and the 910-kev radiation of Y^{89}. This was done successfully with the aid of a sodium iodide-photomultiplier combination coupled to a 50-channel pulse-height analyzer.

A 5-mil niobium foil was bombarded for 1 hour with 100-Mev protons. After dissolving the foil in an $HF-HNO_3$ mixture, LaF_3 was precipitated several times to remove zirconium and yttrium activity. After the last scavenge precipitation an 8-minute growth period was allowed to pass and LaF_3 was precipitated and quickly separated to remove zirconium which had grown in. The LaF_3 was placed in the scintillation spectrometer and data were taken on the gamma spectrum for 1 minute with the purpose of observing the 4.4-minute Zr^{89m} activity, if present. The registers were then reset and data were taken again for 1 minute. Figure 21 shows the series of curves thus obtained. The 288-kev gamma radiation of Zr^{89m} and the 910-kev gamma radiation resulting from the decay of Zr^{89} to Y^{89m} are plainly evident. The activity in the 588-kev peak after correcting for coincidence losses in the mechanical registers decayed with a half life of 4 ± 1 minutes.

After complete decay of Zr^{89m} the gamma spectrum obtained corresponded to the annihilation radiation and the 910-kev gamma radiation expected of

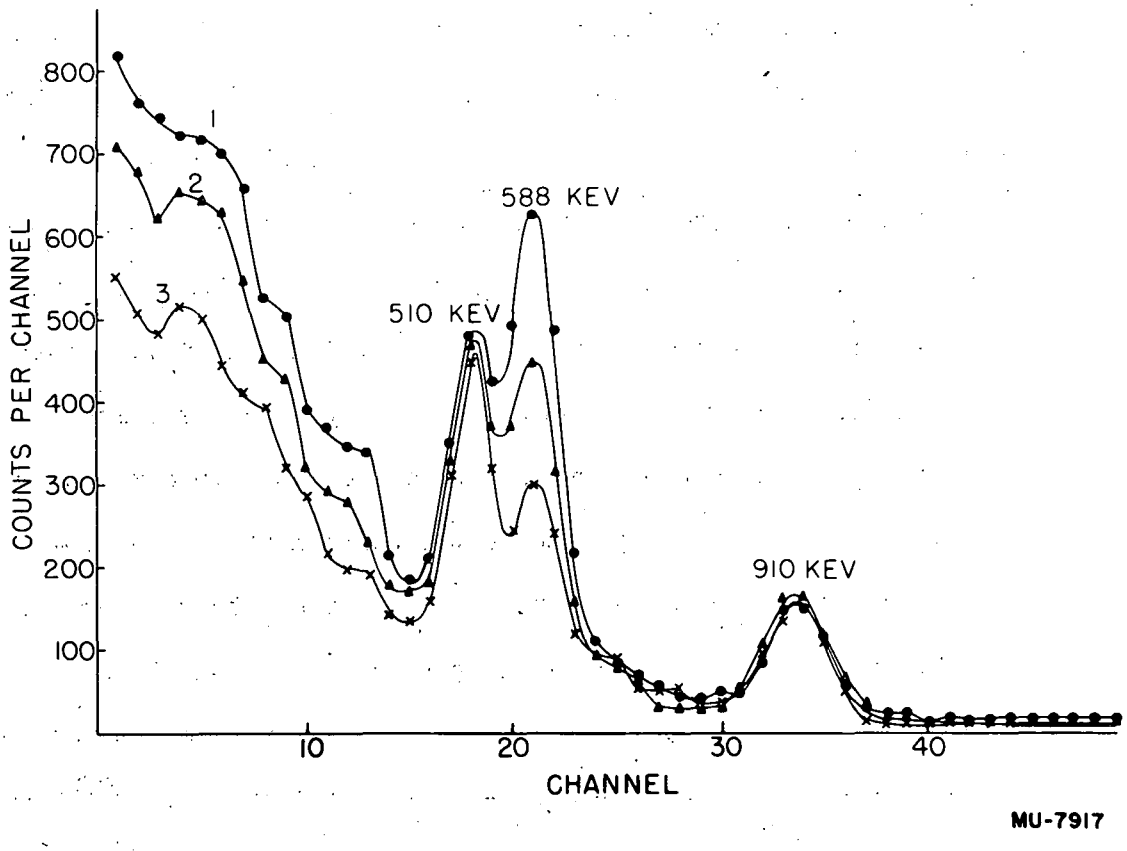

Fig. 21. Gamma spectrum of Zr^{89m} and Zr^{89} isolated from Nb^{89} after an 8-minute growth period. Curve 1 is a 1-minute run started 8.2 minutes after an 8-minute growth period. The 588-kev gamma ray of Zr^{89m} is plainly visible. Succeeding 1-minute runs show the rapid decay of this 4.4-minute activity.

a sample of Zr^{89} (see Fig. 22). This activity decayed with the proper 79-hour half life. Subsequent experiments in which the niobium was given more extensive purification and the zirconium was isolated by precipitation of barium fluozirconate gave the same results.

In a careful series of measurements taken of activity isolated after 2-minute growth periods it was possible to determine the atom ratio of Zr^{89m} and Zr^{89} formed from the decay of niobium. This was done by integrating under the photopeaks and making suitable corrections for half life and counting efficiency. The counting efficiency of the 588-kev gamma ray was taken as 12 percent and the total conversion coefficient as 0.076.[29] The counting efficiency of the 910-kev gamma ray was taken as 7.3 percent and K conversion was considered negligible. The atom ratio obtained was $1.2 \pm 0.5 \times 10^{-2}$, showing that the major decay was going to the $g_{9/2}$ ground state of Zr^{89}. This atom ratio was determined for zirconium daughter fractions isolated after 2-minute growth periods at various times from 1 hour after bombardment up to 8 hours after bombardment. There appeared to be no significant change in the ratio during this time which was taken as an indication that the half life of Nb^{89} $(p_{1/2})$ is close to that of Nb^{89} $(g_{9/2})$.

The whole series of measurements was repeated on a different bombardment of niobium. The same atom ratio within an experimental error of about 30 percent was obtained and this ratio underwent no significant change in a series of milkings covering a 6-hour period after the bombardment.

It was considered desirable to know whether the Nb^{89} $(p_{1/2})$ to Nb^{89} $(g_{9/2})$ ratio was different in a sample prepared by a different method.

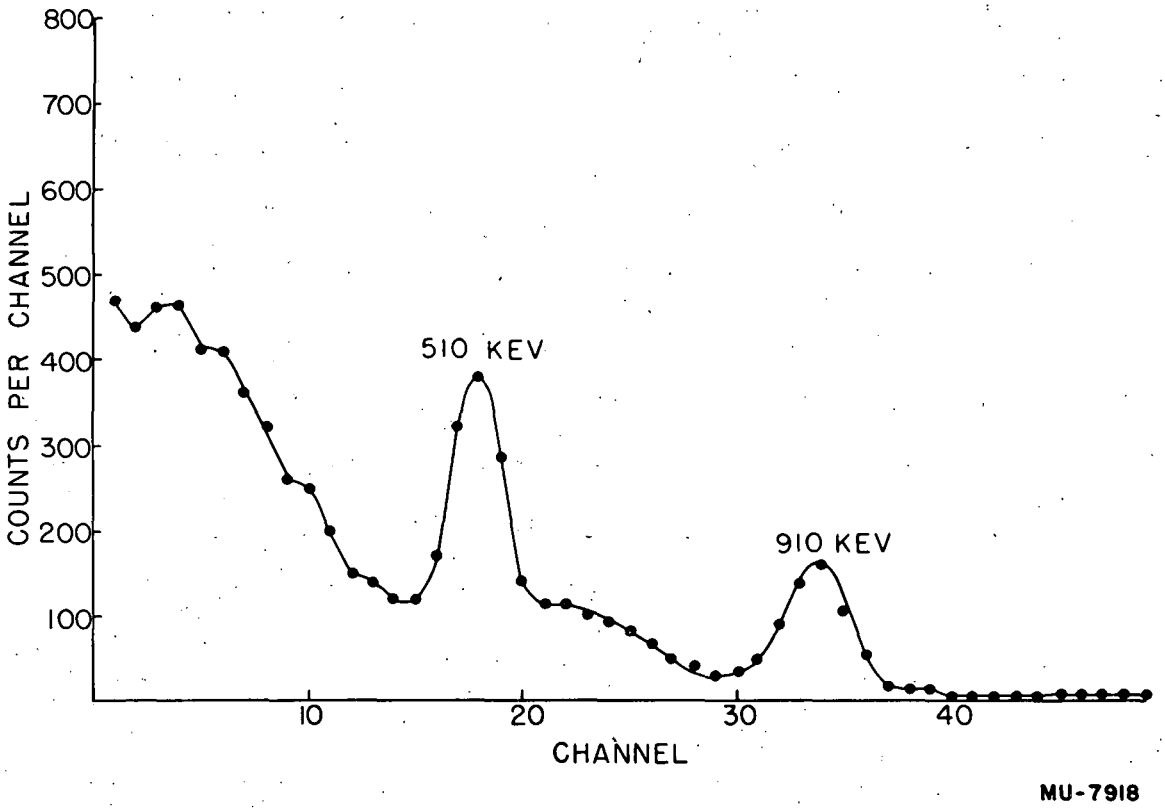

Fig. 22. Gamma spectrum of Zr^{89} daughter activity isolated from initially pure niobium fraction.

For this purpose the niobium fraction of a silver target bombarded with 340-Mev protons was studied. After a 40-minute bombardment the target was dissolved in 10 M HNO_3. Niobium was coprecipitated on MnO_2. This precipitate was dissolved in 12 M HCl and contacted with di-isopropyl ketone to extract the niobium.[30] After niobium was backextracted into water and evaporated to dryness it was taken up in a mixture of 10 M HNO_3 and HF. Zirconium daughter activity was removed periodically by addition of zirconium and barium carrier to precipitate barium fluozirconate. Scintillation spectrometer curves on zirconium fractions so isolated after brief growth periods showed the 588-kev radiation of Zr^{89m}, and the annihilation radiation and 910-kev radiation of Zr^{89}. The atom ratio of Zr^{89m} to Zr^{89} was again observed to be constant for the activity milked at 1-hour intervals for a period of 5 hours. A significant difference, however, in this ratio from that obtained from niobium bombardments was found. In the niobium isolated from silver spallation targets the observed atom ratio was 3×10^{-3}, or a factor of 4 lower. This difference is believed to be well outside the experimental error.

As a further check the same ratio was measured in a similar manner on a niobium sample prepared by bombardment of zirconium-metal foil with the 32-Mev proton beam of the Berkeley linear accelerator. In this case an atom ratio of about $2.6 \pm 0.5 \times 10^{-2}$ was obtained which is a factor of approximately 2 greater than in the samples isolated from niobium targets and a factor of about 9 greater than in the silver bombardment case.

The gamma spectrum of the purified niobium fraction was run at a time when more than 3/4 of the activity was Nb^{89}, and on samples purified 24 hours later when Nb^{90} was the chief activity. A comparison of the two,

spectra showed that Nb^{89} did not emit any gamma rays in addition to the annihilation radiation and the only gamma rays in both spectra were those of Nb^{90} reported by Boyd and Ketelle.[31] The 588-kev gamma ray was not intense enough in comparison with the 510-kev annihilation radiation peak for it to be observed. If Nb^{89m} decayed chiefly by isomeric transition, a prominent gamma ray of less than 300 kev should have been observed.

During the course of these studies considerable data on the radiations of Nb^{90} were obtained. This will be reported in Section V.

The positron endpoint energy was determined by using an anthracene crystal-photomultiplier combination coupled to a 50-channel pulse analyzer. Figure 23 shows a typical endpoint determination of 2.85 ± 0.10 Mev for the $Nb^{89} \rightarrow Zr^{89}$ transition. It was established that these positrons decayed with a 2-hour half life.

B. Carbon Ion Bombardments

In addition to the bombardments mentioned above Nb^{89} was prepared in a novel way by the bombardment of bromine with accelerated carbon ions. This was done in the 60-inch cyclotron using a beam of hextuplicately charged carbon ions.[32] The carbon ions in the beam have a continuous spread of energy with a maximum energy of 120 Mev. Sodium bromide powder wrapped in thin tantalum foil was bombarded to carry out the reactions:

$$_{6}C^{12} + {}_{35}Br^{81} \rightarrow {}_{41}Nb^{90} + 3{}_{0}n^{1}$$

$$_{6}C^{12} + {}_{35}Br^{81} \rightarrow {}_{41}Nb^{89} + 4{}_{0}n^{1} .$$

After bombardment the sodium bromide was dissolved in 10 M HNO_3 and extracted with CCl_4 to remove bromine. The MnO_2 was precipitated to remove

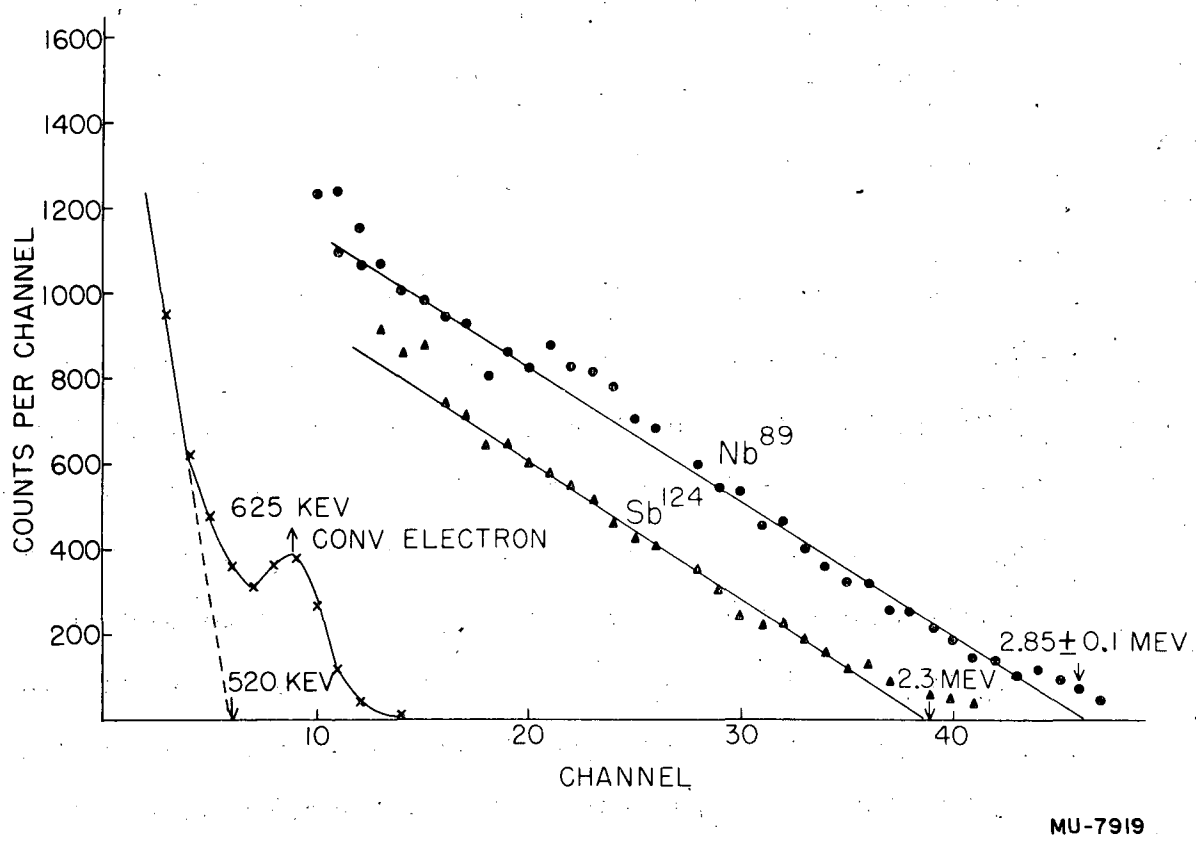

Fig. 23. Positron endpoint of Nb^{89} as determined on anthracene crystal spectrometer. Standards used were Cs^{137} and Sb^{124}.

niobium. The MnO_2 was dissolved in 12 M HCl and from this solution niobium was extracted with di-isopropyl ketone. Backextraction of the niobium into water completed the purification. Resolution of a GM-decay curve extrapolated back to the end of the bombardment showed 5/6 of the activity to be 1.9-hour Nb^{89} and 1/6 to be 14.6-hour Nb^{90}. The atom ratio, assuming equal counting efficiencies, is 0.7. Gamma analysis within 2 hours of the end of the bombardment showed the gamma rays of Nb^{90} and no others. Gamma analysis of zirconium daughter fractions showed the 910-kev gamma ray of Y^{89m}. No attempt was made to search quickly for Zr^{89m}.

C. Discussion

The experimental data are best summarized by the decay scheme of Fig. 20. This scheme is incomplete because the energy separation of the Nb^{89} states, and the identity of the higher state, has not been determined. The small ratio of the Nb^{89m} to Nb^{89} in the samples we were able to prepare made it impractical to get the energy separation by the resolution of the positron spectrum into two components. From the systematics of the $p_{1/2}$-$g_{9/2}$ separation quoted previously[5] it is expected that this separation in Nb^{89} is quite small and hence the isomeric transition branching must be slight. By preparing very active samples and looking for the electrons of a highly converted M4 transition in the 0- to 200-kev range with a precision spectrometer it might be possible to learn this energy separation.

Figure 24 differs from a very similar scheme presented by Diamond,[33] who independently has carried out a study of these isomers, only in that he assigns a half life of 0.8 hour to the $p_{1/2}$ level. This was done on

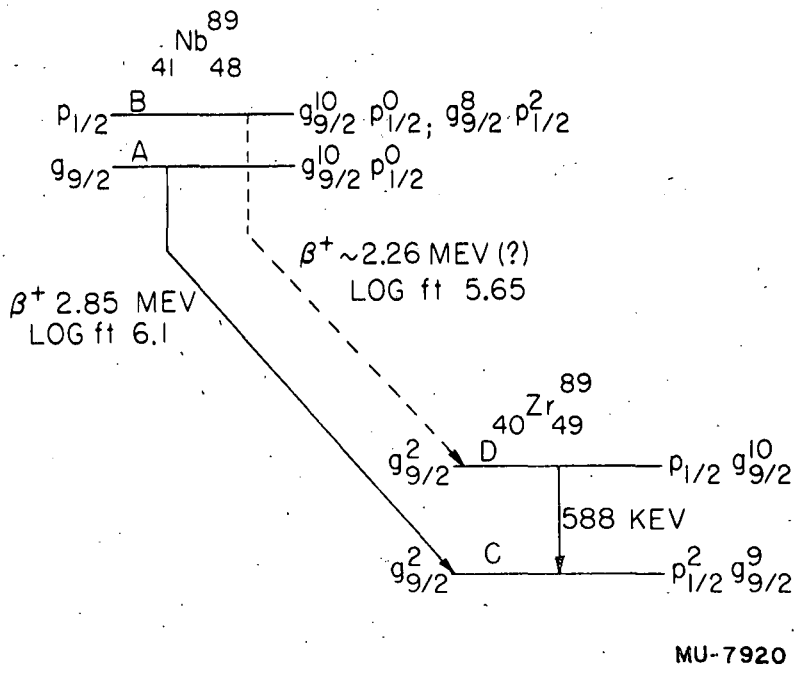

Fig. 24. Decay scheme for Nb^{89} showing the main configurations of the proton (left) and neutrons (right). The $g_{9/2}$ proton in A stabilizes the even-even core in a $g^{10}_{9/2}$ configuration. The transition AC is delayed because of the necessity of rearrangement of the neutron core to produce two $p_{1/2}$ neutrons in C. B is indicated with a mixed neutron configuration, but the $g^{8}_{9/2}$-$p^{2}_{1/2}$ configuration probably predominates. Hence the transition BA would be delayed.

the basis of a series of quantitative timed milkings of zirconium daughter activity carried out in much the same manner as described in this report except that the determination of the relative amounts of 4.4-minute Zr^{89m} and 78-hour Zr^{89} activity was done by the resolution of a decay curve taken on a scintillation counter without pulse-height analysis. Because our data are based only on the activity in the 588- and 910-kev gamma peaks we feel they are somewhat more trustworthy.

As mentioned above the atom ratio of Nb^{89} $(p_{1/2})$ to Nb^{89} $(g_{9/2})$ was constant over a period of several hours for niobium samples prepared from niobium, silver and zirconium targets. This is inconsistent with the half life of 0.9 hour reported by Diamond for Nb^{89} $(p_{1/2})$. It is not clear why the two methods give different results, although a slight constant contamination of the zirconium milkings with improperly resolved niobium activity could account for it.

From Moszkowski's[34] graphs the log ft value for the 2.85-Mev positron transition Nb^{89} $(g_{9/2}) \rightarrow Zr^{89}$ $(g_{9/2})$ is 6.1 which is considerably higher than that expected (5.0 ± 0.3) for an allowed transition. If the unknown Nb^{89m}-Nb^{89} separation is neglected, the log ft value of the Nb^{89m} $(p_{1/2}) \rightarrow Zr^{89m}$ $(p_{1/2})$ transition is 5.65 which is still somewhat higher than expected. These two transitions can be added to a growing list of apparent exceptions to the usual rules of beta decay discussed by de-Shalit and Goldhaber.[35] For example, the well-established Zr^{89} $(g_{9/2}) \rightarrow Y^{89m}$ $(g_{9/2})$ transition has a log ft value of 6.1 and similarly, the transition Zr^{89m} $(p_{1/2}) \rightarrow Y^{89}$ $(p_{1/2})$ has a log ft value of 6.85. These and other examples are discussed by de-Shalit and Goldhaber[35] who give an explanation in terms of a rearrangement of nucleons in the even-even core of an odd-A nucleus.

Following their development the decay scheme of Fig. 1 can be redrawn in the form of Fig. 24.

IV. STUDIES ON Mo^{90} AND THE NEW ISOMERS Nb^{90m_1}, Nb^{90m_2}

The persistence of islands of isomerism near the closed shell of 50 or 82 particles irrespective of the fact whether they are odd-A or even-A nuclei, suggests that the even-A isomers are also likely to occur in the same region as odd-A isomers because to a first approximation the individual configurations of the proton or neutron are not influenced by one another. In this section isomerism in the odd-odd nucleus Nb^{90} will be discussed.

Diamond[36] at Harvard University had studied Mo^{90} prepared by proton bombardment of niobium. He reported Mo^{90} to be a positron emitter with a half life of 5.7 ± 0.2 hours. The mass assignment was made by identification of the 14.6-hour Nb^{90} formed in its decay. He reported positrons of maximum energy of 1.4 Mev and gamma rays at energies of 1.0, 0.24 to 0.26, and 0.1 to 0.13 Mev. The Mo^{90} has 42 protons and 40 neutrons and decays by positron emission to Nb^{90} which has 41 protons and 49 neutrons. According to the shell model the odd proton and the odd neutron in Nb^{90} may exist in the state $g_{9/2}$ or $p_{1/2}$, suggesting thereby the possibility of an isomeric state. With this purpose in view, a detailed study of Mo^{90} and the 14.6-hour Nb^{90} was undertaken. During the course of this work it was indeed found that Nb^{90} exists in isomeric states; in fact three such states were established.

A. Molybdenum 90

In our experiments with Mo^{90} produced by the bombardment of Nb^{93} with 80-Mev protons in the 184-inch cyclotron, we found Mo^{90} to have positrons of maximum energy of 1.15 ± 0.1 Mev. Figure 25 shows the positron end-point as determined by an anthracene crystal spectrometer.

Figure 26 shows the gamma spectrum of Mo^{90} as determined by a sodium iodide crystal-photomultiplier assembly coupled to a 50-channel differential-pulse analyzer. There are two gamma rays of 120- and 250-kev energy in addition to the annihilation radiation. The small peak at 65 kev is due to the platinum x-rays produced by the platinum backing on which the sample was mounted. Using the values given by Kahn and Lyon[37] for the counting efficiency as a function of energy for a sodium iodide crystal 1.5 inches in diameter and 1-inch thick, we assumed a counting efficiency of 100 percent for 120-kev gamma rays and 60 percent for 250-kev gamma rays and found the two gamma rays to be in almost equal abundance. This calculation does not allow for conversion which, as will be seen, is appreciable. No 1-Mev gamma radiation was detected in a freshly prepared sample of Mo^{90}, although a gamma ray of 1.14 Mev did appear later as Mo^{90} decayed to Nb^{90}.

Conversion electrons of the 120- and 250-kev gamma rays were also measured by a magnetic double-focusing spectrometer. Figure 27 shows the spectrum thus obtained. The K/L conversion ratios of 120- and 250-kev gamma rays were determined to be 3.6 ± 0.2 and 5.2 ± 0.2, respectively.

Gamma-gamma coincidence studies were carried out with Mo^{90} with the following results. With 510-kev annihilation radiation or 16.5-kev x-rays at the gate, no coincidences were observed with 120- or 250-kev

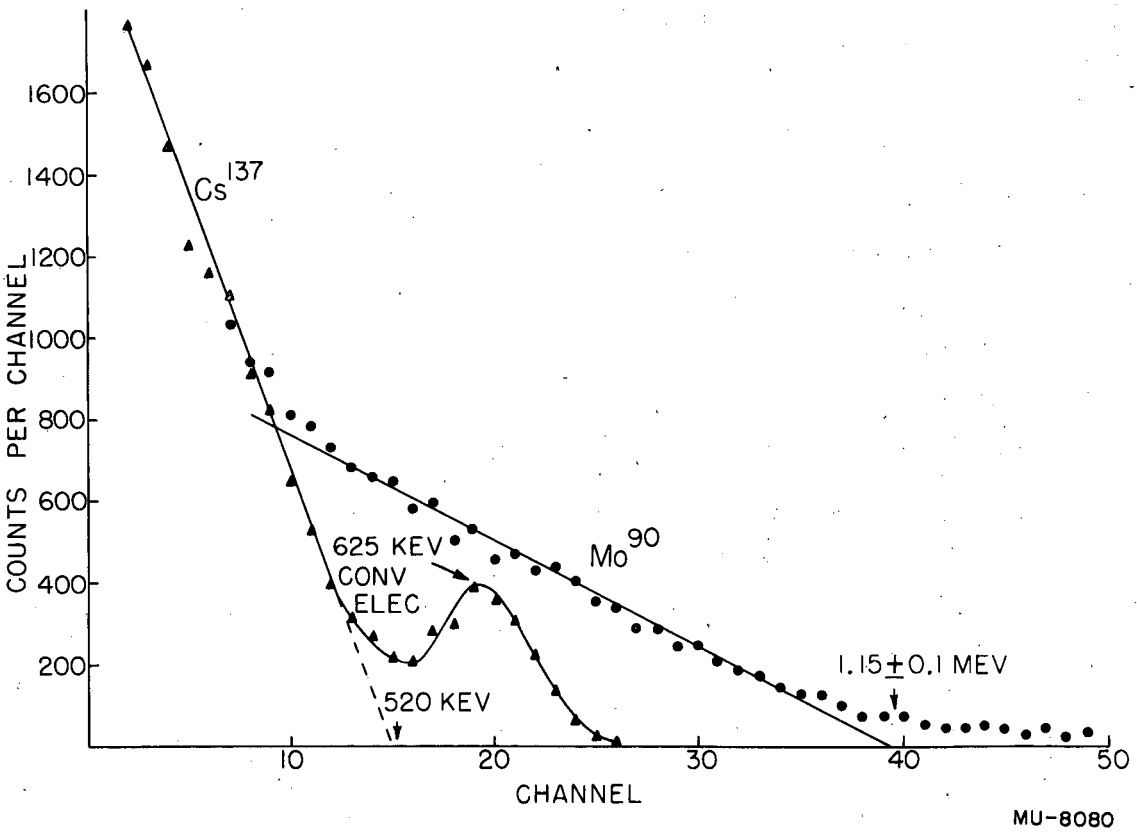

Fig. 25. Positron endpoint of Mo^{90} as determined on an anthracene crystal spectrometer. Cs^{137} was used as a standard.

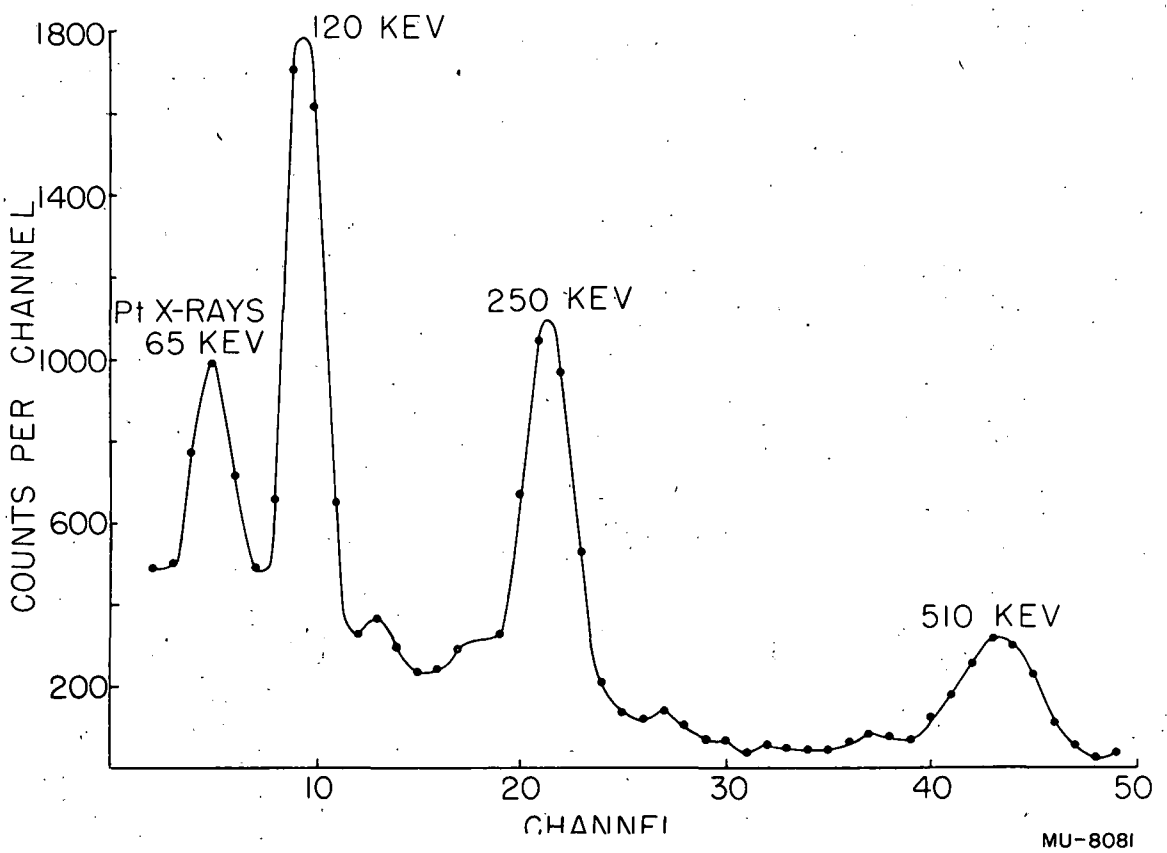

Fig. 26. Gamma spectrum of Mo^{90}.

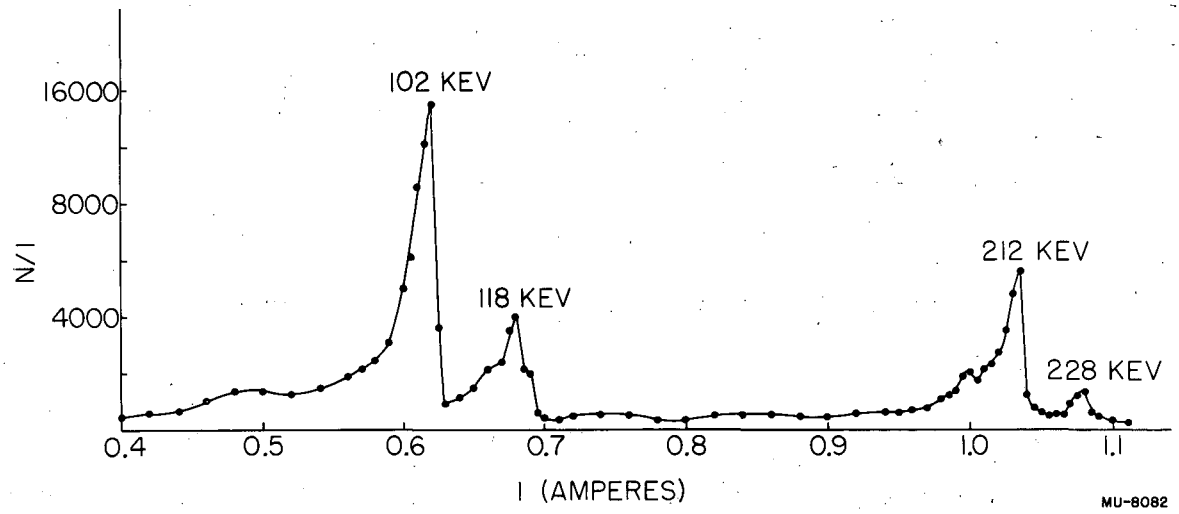

Fig. 27. Conversion electrons of Mo^{90}.

gamma rays. Similarly, by using 120-kev gamma rays, no coincidences were observed with either the 250-kev gamma ray or the annihilation radiation. Likewise we obtained no coincidences with the 120-kev gamma ray or the annihilation radiation when the 250-kev gamma ray was put at the gate. The nonexistence of coincidences between 510-kev annihilation radiation or 16.5-kev niobium x-rays and the two gamma rays of 120- and 250-kev indicates that both the gamma rays are delayed.

B. The 24-Second Isomer, Nb^{90m_1}

The gamma-gamma coincidence studies suggested the existence of an isomeric state of Nb^{90} in addition to the ground state of Nb^{90}--the 14.6-hour activity previously studied by Boyd,[38] Kundu and Pool,[39] and Jacobson and Overstreet.[40] Experiment soon showed that a 24 ± 3 second activity was formed in the decay of Mo^{90}. An intense carrier-free sample of Mo^{90} was placed on the top shelf of our scintillation counter for a 1-minute growth period. The aluminum front cover on the sodium iodide crystal was not grounded and it carried a potential of 500 volts negative with respect to the sample. The sample was then quickly withdrawn and the recoil daughter atoms of Nb^{90m_1} and Nb^{90} collected from the molybdenum source during the growth period were counted. The gamma spectrum was determined by a 50-channel differential-pulse analyzer. Figure 28 shows the gamma spectrum of the recoil activity immediately after the growth period. The gamma spectrum of Mo^{90} is also shown in the same figure for comparison. The readings of the register at the peak of the 120-kev gamma ray were taken every 0.2 minute for a 2-minute period without disturbing the count switch and from the analysis of the data a

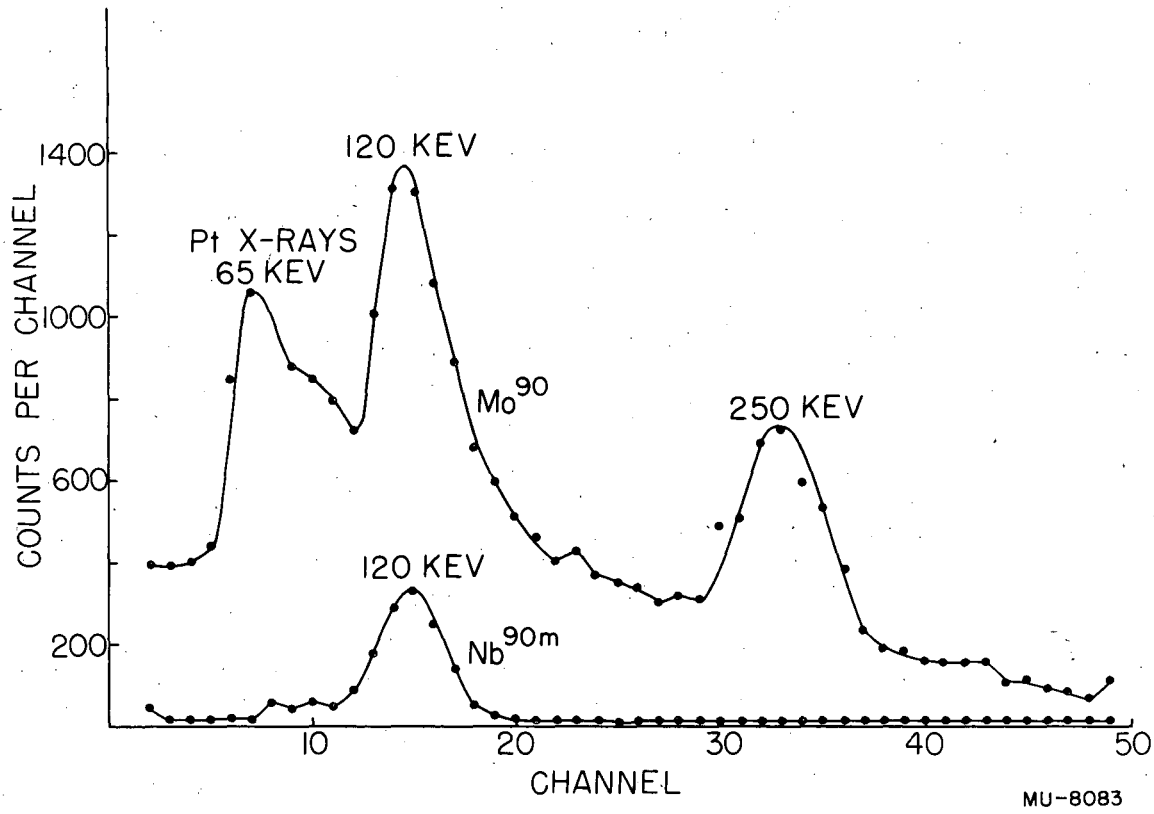

Fig. 28. Gamma spectrum of Nb^{90m}. The gamma spectrum of Mo^{90} is also shown for comparison.

half life of 24 ± 3 seconds was obtained for the recoil activity (see Fig. 29).

The fact that this 24-second activity is the product of the decay of Mo^{90} was further confirmed by plotting the yield of the recoil activity in a 1-minute growth period as a function of the time at which the milking was done. A half life of 5 hours was obtained for the molybdenum parent as shown in Fig. 30, confirming thereby the genetic relationship.

The 24-second isomer was proved to be a niobium activity in the following manner. From the work of Huffman et al.,[41] Kraus and Moore,[42] and Hicks, Stevenson and Gilbert,[43] it was known that niobium and molybdenum form complexes in strong HCl which are strongly adsorbed on anion-exchange resins. On reinvestigation it was found that a mixture of molybdenum and niobium in 6 M HCl is adsorbed by Dowex-1, a strong-base quaternary amine polymer. Niobium can be eluted with 5 M HCl but molybdenum is strongly held at this HCl concentration. The Mo^{90} activity in 6 M HCl solution was run through a 2- x 0.5-cm column of Dowex-1 previously equilibrated with 12 M HCl. The molybdenum on the column was washed free of niobium activity with 5 M HCl and, after a growth period of 1 minute, niobium formed by decay during this period was quickly stripped from the column by 5 M HCl, collected in a 2 ml lusteroid cone and studied in a scintillation counter for the determination of the gamma spectrum. The spectrum showed the presence of 120-kev gamma rays decaying with a half life of 24 ± 3 seconds, thereby further establishing that the 24-second activity was a niobium activity.

If the 24-second half life and the 120-kev energy are substituted into the Montalbetti[10] nomogram of Weisskopf's formula the predicted half lives for various type transitions are $\sim 10^{-6}$ second for E2, $\sim 2 \times 10^{-4}$ second for

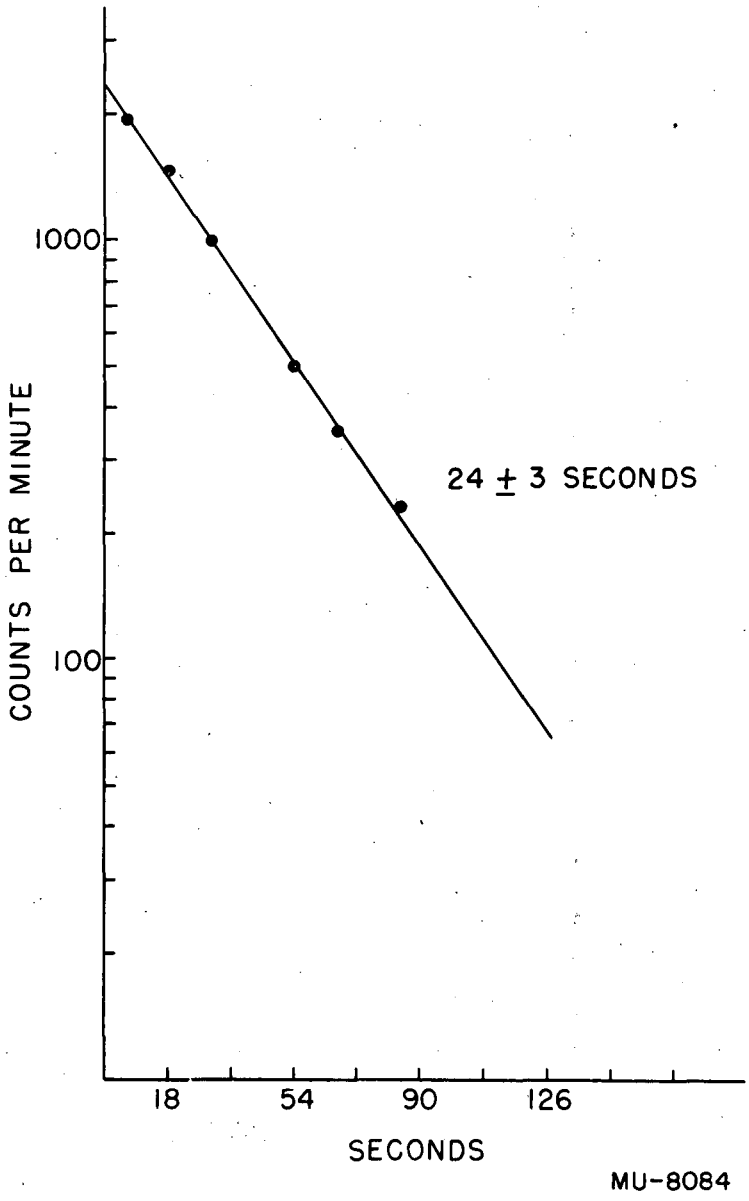

Fig. 29. Decay of gamma ray of 120-kev in Nb^{90m1}.

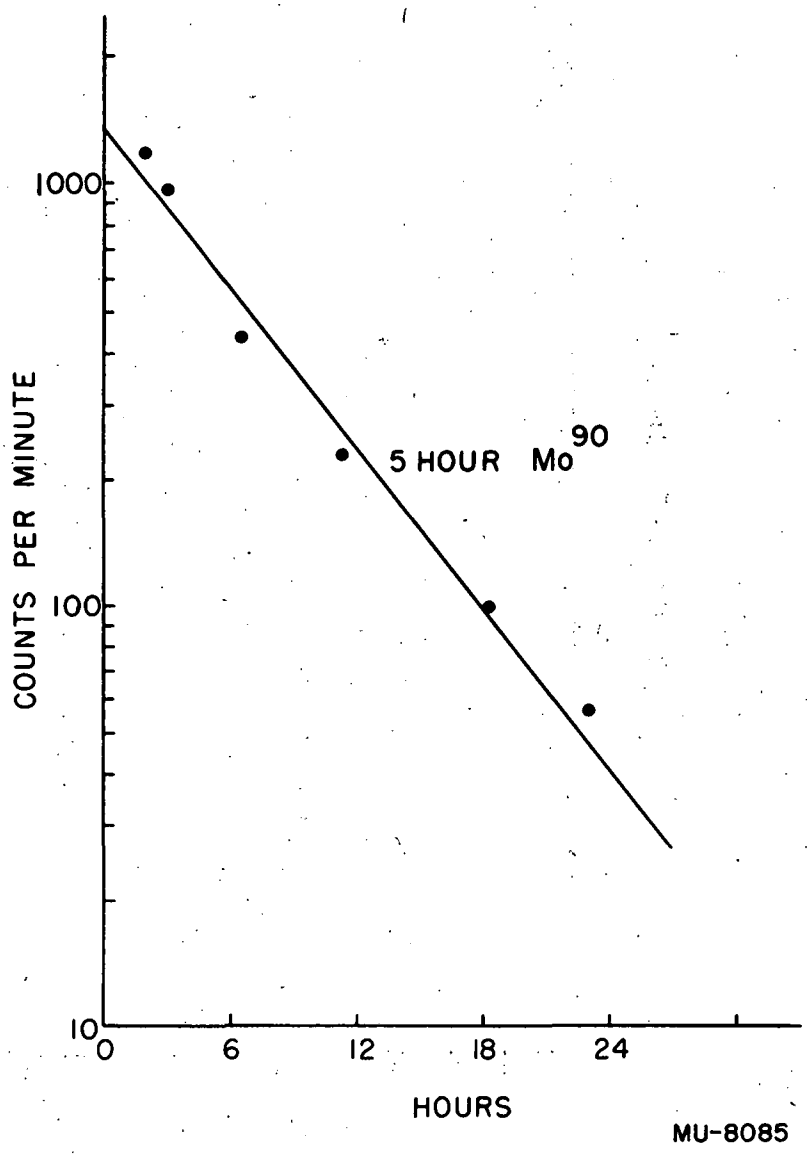

Fig. 30. Yield of 24-second Nb^{90m} from timed milkings of Mo^{90} parent sample.

M2, 2 seconds for E3, 200 seconds for M3, ~100 days for E4 and ~30 years for M4. From this it seems clear that the multipole order is 3.

When this multipole assignment was checked by the K/L ratio method and the K-shell conversion coefficient method discussed in the introduction, however, the results were anomalous. The observed K/L ratio is 3.6, which is higher by a factor of about 2 than the empirical curves of Goldhaber and Sunyar would suggest for an E3 transition and in closer agreement with the predictions for an E2 transition. The K/L empirical curve for M3 transitions is poorly defined but the prediction would be a value of about 5.

A more serious discrepancy arises in connection with the K-conversion coefficient. The curves of Rose et al.[11] do not provide values of conversion coefficients for gamma energies below 150 kev. An extrapolation of their curves for Z = 40 indicates that the expected K-shell conversion coefficient for an E3 transition is of the order of 2 to 4 and somewhat higher for an M3 transition. An experimental measurement of the K-shell coefficient for the 120-kev gamma ray gave a value of 0.5, far out of line with the theoretical value and much closer to the value expected for E2 transitions.

The experimental measurement was carried out in this manner. A sodium iodide crystal covered with a beryllium window was substituted for the usual aluminum covered crystal in the gamma-ray spectrometer. Using this crystal, a sample of Mo^{90} was examined for K x-rays of 16.5-kev energy. A gamma spectrum was taken with the gain settings so selected that the 120-kev gamma peak also appeared on the same scale. After correction for absorption losses and fluorescence yield in the x-ray peak the ratio of the K vacancies to the number of 120-kev quanta was calculated.

The absorption loss was estimated by using the same counter to determine the L x-ray spectrum of a sample of Am^{241}. The area under the unresolved 17.3- and 13.5-kev L x-ray peaks was compared with the area under the 59.9-kev gamma peak. The true relative abundances were taken from the work of Beling, Newton and Rose.[44] A calculation showed that for this particular L x-ray mixture the absorption loss was about 25 percent in the beryllium window and magnesium oxide reflector around the crystal. It was assumed that the absorption loss of the 16.5-kev x-rays of niobium was the same. This is not strictly correct but the error cannot be more than 50 percent.

The fluorescence yield was estimated as 0.73 from the tables of Broyles, Thomas and Haynes.[45]

With these corrections and assumptions the ratio of K-shell vacancies to 120-kev quanta was calculated to be 1.5. This cannot be taken as the conversion coefficient since some of the K rays may be accounted for by K capture, or by conversion of the 250-kev gamma ray, but it can be taken as an upper limit. A more significant measurement was made by collecting some of the 24-second activity by recoil on the case of the crystal in the manner discussed above and measuring the low-energy spectrum of this activity. The gamma spectrum of the recoil activity showed that the larger part of the x-ray peak observed in the Mo^{90} sample has to be assigned to K capture. The estimate of the K-shell conversion coefficient of the 120-kev gamma ray is ~0.5.

This is a real anomaly which has not been resolved. The conversion coefficient is a factor of 4 to 8 too low for a transition of multipole order of 3, while the half life and energy are in agreement only with a spin change of this value.

C. Preliminary Half-Life Studies on 250-kev Isomeric State Nb^{90m2}

From our gamma-gamma coincidence studies it was concluded that both the 120- and 250-kev gamma rays were delayed. Because the 250-kev gamma ray did not occur in the gamma spectrum of the 24-second isomer, it is evident that the 250-kev level does not lie below the 120-kev level.

It may be that the main positron decay of Mo^{90} goes to a level in Nb^{90} which deactivates by a 250- to 120-kev cascade.

Chemical separation of niobium from Mo^{90} showed only the presence of 120-kev gamma rays of 24-second half life and therefore the half life of the 250-kev state is not more than 24 seconds. By performing the chemical separation rapidly, and from further recoil studies, we estimated the half life of the 250-kev transition to be less than a few seconds. Because we were not able to observe any coincidences of 250-kev gamma rays with either 120-kev gamma rays or the annihilation radiation, the half life of the 250-kev gamma rays should be greater than 5 microseconds, the resolving time of the coincidence circuit.

Making use of Montalbetti's[10] nomogram, the Weisskopf predictions of half lives for transitions of various types are as follows: $\sim 10^{-8}$ second for E2, 10^{-6} second for M2, 10^{-2} second for E3, 1 second for M3, 10 hours for E4, and 30 days for M4. Hence again the transition type is narrowed to M3 or E3.

After establishing the delay in the 250-kev tansition to be between 5 microseconds and 1 second, a novel device was used to narrow the limit to less than 0.1 second. A carrier-free sample of Mo^{90} mounted on a platinum plate was supported on a stand with its face at a distance of about 0.5 cm from a moving tape of aluminum or paper. A potential

difference of 900 volts was applied between the tape and the sample, the negative terminal of the battery being connected to the tape. Those few recoiling atoms of niobium formed in the decay of Mo^{90} which escaped from the source were collected on this tape and were transported through a scintillation counter. The gamma spectrum was studied by means of the 50-channel differential-pulse analyzer. The time taken to transport the recoil atoms of niobium on the tape to the scintillation counter could be varied by changing the speed of the driving motor. With the slowest speed it took nearly 5 seconds for the recoils to reach the scintillation counter which was at a distance of 1 foot from the sample, while at the fastest speed of the motor the time taken was about 0.1 second. When the tape was moved at the slowest speed for 1 minute we obtained a peak at 120 kev in the gamma spectrum but there was no indication of any gamma ray of 250-kev energy. By increasing the speed the number of total counts under the 120-kev peak was reduced. It is therefore possible to discriminate in favor of a shorter half life. No indication of 250-kev radiation was gotten even by increasing the speed to the maximum and it was concluded that the delay in the 250-kev transition should be less than 0.1 second and more than 5 microseconds.

The overall efficiency for collection of recoils by this method is quite small (<0.01 percent) but by using very intense carrier-free sources there was no difficulty in collecting a few hundred counts per minute of the 24-second isomer and hence there should have been no trouble in collecting enough of the 250-kev state for identification if its half life were greater than 0.1 second.

D. Measurement of Half Life of Nb^{90m_2}

At this point the half life of Nb^{90m_2} was considered to be narrowed to the region 0.1 second to 5×10^{-6} second. From the theoretical predictions it seemed most likely that the half life should fall in the millisecond region. Hence an electronic delayed-coincidence technique was used to search the millisecond region. This equipment was assembled by Mr. A. E. Larsh. Gamma radiation from a weak sample of Mo^{90} with a count rate of about 50 counts per minute was detected by a sodium iodide crystal. Annihilation radiation was selected by a single-channel pulse-height analyzer and the output of the analyzer was fed to the trigger input of a Tektronix 514D oscilloscope to start a trace across the oscilloscope face. A second sodium iodide crystal was simultaneously detecting gamma radiation from the Mo^{90} sample. These signals, after amplification, were fed to the vertical deflecting system of the oscilloscope. Signal pulses arriving in prompt coincidence with the annihilation trigger pulses were displayed as dots at the start of the trace. The energy of the gamma radiation was proportional to the amount of vertical deflection on the face of the cathode ray tube. Any delayed gamma ray signal pulses or randomly occurring chance pulses which arrived during the time it took the trace to traverse the oscilloscope face appeared as dots at a corresponding distance from the start of the trace.

A Poloroid Land camera was used to take 20-minute exposures of the scope face. A finished photograph showed three bands of dots. The density of dots corresponding to chance pulses of 120- and 510-kev radiation was small and did not decrease across the trace. The density of dots corresponding to 250-kev radiation showed a real decrease to a chance background of 25 to 35 percent of the initial density when the trace speed was set at 1 cm per 4.5 milliseconds.

The density of dots in the 250-kev band was plotted on the logarithmic scale of semilogarithmic graph paper versus the distance (time) from the beginning of the traces. The chance background was subtracted to show the true decay curve. Analysis of many photographs gave a value of 10 to 20 milliseconds for the half life of the 250-kev radiation.

In initial trials of this method halatron from the base line made the recording of the desired data almost impossible. Hence some changes from the normal operation of the Tektronix instrument were required. The normal unblanking circuit was not used. Instead, when the positive signal pulse was fed to the vertical deflection system as mentioned above it was simultaneously fed to an inverter circuit which inverted and amplified the pulse. The resulting negative pulse was fed to the cathode input post on the oscilloscope to unblank the trace at the instant a signal appeared on the vertical amplifier.

The tables of Rose et al.[11] give precise values of 0.14 and 0.33 for the K-shell conversion coefficient for an E3 and an M3 transition, respectively. It was not possible to estimate this coefficient experimentally by the method used in the case of Nb^{90m_1}.

V. THE RADIATIONS OF 14.6-HOUR Nb^{90} AND AN ISOMERIC STATE IN THE EVEN-EVEN NUCLIDE Zr^{90}

The 14.6-hour ground state of Nb^{90} produced as the end product of the decay of Mo^{90} was studied by several workers. Boyd[38] reported Nb^{90} to be a positron emitter with a maximum energy of approximately 1.7 Mev, while Diamond,[36] and Kundu and Pool[39] have reported the positrons to have a maximum energy of 1.2 Mev. Boyd and Ketelle[31] reported three gamma rays of energies 140 kev, and 1.14 and 2.23 Mev in Nb^{90}.

A redetermination of the positron endpoint of Nb^{90} by means of an anthracene crystal spectrometer coupled to a 50-channel differential-pulse analyzer yielded a value of 1.7 ± 0.1 Mev for the positron endpoint as shown in Fig. 31. The gamma spectrum of Nb^{90} is shown in Fig. 32. There are three gamma rays of energies of 140 kev, 1.14 and 2.20 Mev in addition to the annihilation radiation. Taking into account the counting efficiency of the sodium iodide crystal for the three energies, the ratios of their abundances are 1/2.8/1.7.

Coincidence measurements made on the gamma rays of Nb^{90} showed that the 140-kev and 1.14-Mev gamma rays are in coincidence with each other and with the annihilation radiation. From the quantitative data the indication is that the 140-kev transition lies above the 1.14-Mev gamma ray. This is in keeping with the expectations from the systematics of the first excited state of even-even nuclei since in Zr^{90} one would expect the first excited state to be greater than 1 Mev.[17] The 2.20-Mev gamma ray showed no coincidences with 140-kev or 1.14-Mev gamma rays or the annihilation radiation.

This result suggested the possibility of a delayed transition involving the 2.20-Mev level in Zr^{90}, although the possibility existed that this level was reached by K-capture decay rather than positron emission. This possibility was eliminated by showing that the intensity of K x-rays relative to the 140-kev peak was only about 80 percent and hence too low by a factor of 2 to account for the 2.2-Mev radiation. The x-ray intensity measurement was made with the sodium iodide crystal with beryllium window described above. Furthermore, a coincidence experiment in which K x-rays were applied to the gate showed no evidence of an x-ray-2.2-Mev gamma ray coincidence.

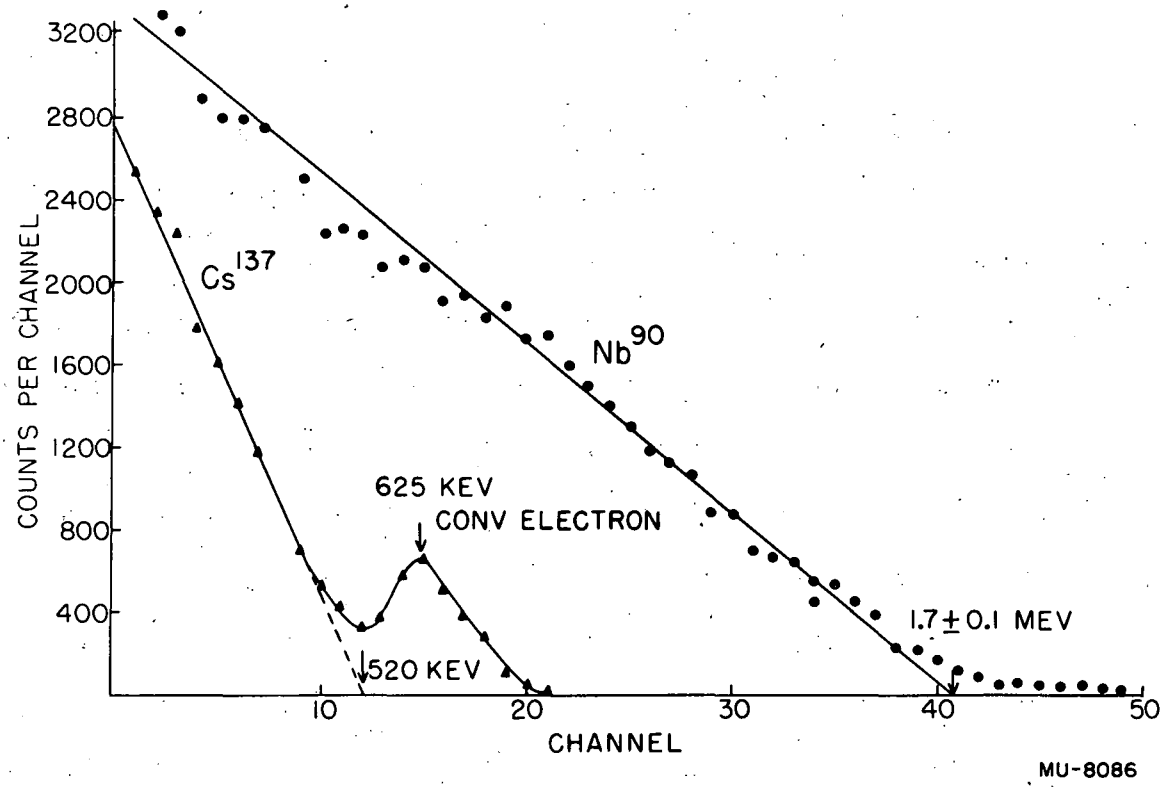

Fig. 31. Positron endpoint of Nb^{90} as determined in an anthracene crystal spectrometer. Cs^{137} was used as a standard.

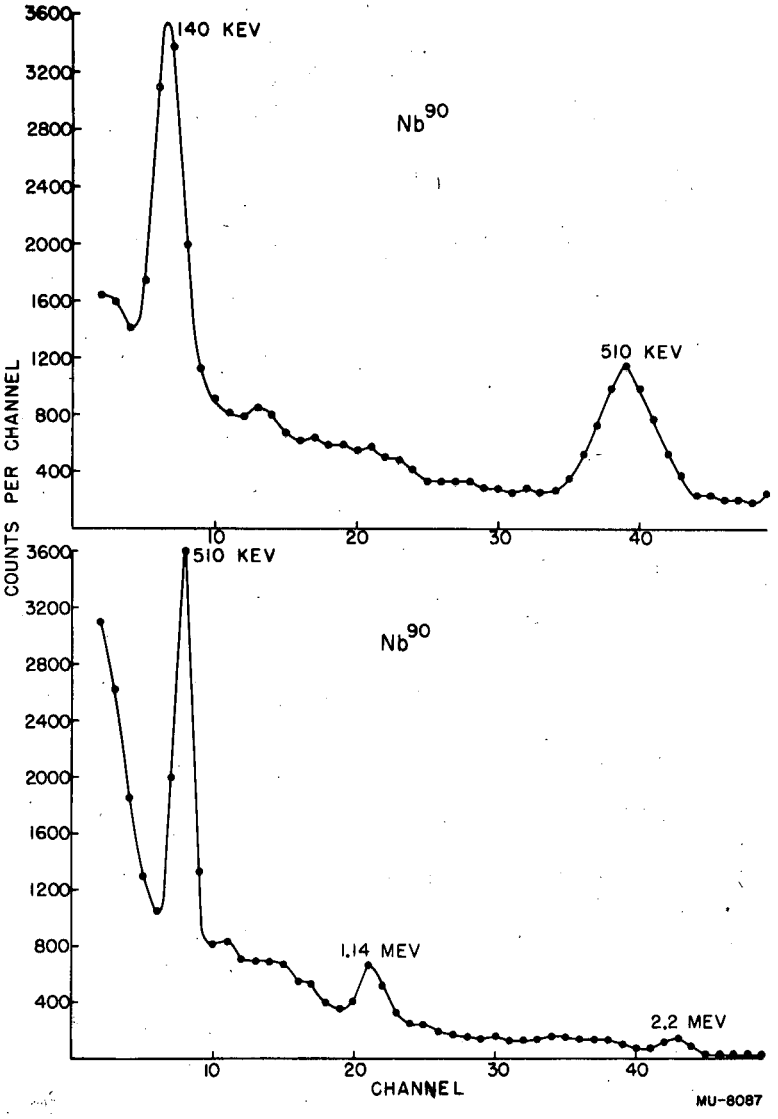

Fig. 32. Gamma spectrum of Nb^{90}.

From a Nb^{90} sample zirconium was precipitated as barium fluozirconate and did not show any gamma ray of 2.20 Mev in its gamma spectrum. By performing this separation rapidly, and by immediate counting of the resulting barium fluozirconate precipitate in a scintillation counter, the delay was fixed as less than 1 minute.

As discussed in the introduction, delayed states are quite rare in even-even nuclei. It would be desirable to determine more fully the characteristics of this new example. Considerable thought was given to the problem of determining the half life by some instrumental method but no feasible way could be devised with the equipment available. The low-counting efficiency of the 2.2-Mev radiation in the 1.0 x 1.5-inch sodium iodide crystals is a serious disadvantage.

Half-life predictions from the Montalbetti[10] nomogram are as follows: 10^{-13} second for E2, 10^{-11} second for M2, 2×10^{-9} second for E3, 2×10^{-7} second for M3, 8×10^{-5} second for E4, 8×10^{-3} second for M4, 1 second for E5, and 100 seconds for M5. Because the resolving time of the coincidence equipment was only about 5×10^{-6} second and an upper limit of 60 seconds was set by chemical experiments, the transition type is narrowed to E4, M4 or E5.

VI. STUDIES ON THE NEUTRON-DEFICIENT ISOTOPES Xe^{123}, Xe^{122}, AND Xe^{121} AND THE IODINE DAUGHTERS FORMED IN THEIR DECAY

Momyer and Hyde,[46] of this laboratory, during the course of a study of isotopes of emanation had made use of the glow-discharge method to deposit the emanation activities on a metallic wire or foil. The radioactive samples prepared in such a way could be studied in a manner similar to

that used for nongaseous samples. This technique was so convenient that it was thought desirable to exploit its use further by undertaking a study of other gaseous activities. The glow-discharge method, which is described in some detail in a later section, has been applied with equal success to the study of xenon activities. During the course of this work some information has been collected on the nuclear properties of some new neutron-deficient isotopes of xenon and the iodine daughters produced in their decay.

This section deals with the nuclear properties of Xe^{123}, Xe^{122} and Xe^{121}, and the iodine daughters formed by their decay. Isolation and study of these iodine isotopes revealed the presence of the known iodine isotopes of masses 123, 122 and 121. Timed separations of the iodine daughter isotopes established the genetic relationship of these to 19-hour Xe^{122}, 1.8-hour Xe^{123} and ~40-minute Xe^{121}. Because our results agree with A. E. Tilley[47] at McGill University and B. Dropesky and E. O. Wiig[48] at the University of Rochester, we shall not give complete details of this part of the work. The purpose of this report is to record some unpublished observations of the radiations of these isotopes carried out with a scintillation spectrometer and a beta-ray spectrometer.

A. Xenon 122 and Iodine 122

Potassium iodide targets were bombarded for 1 hour with 100-Mev protons. The xenon fraction was isolated 24 hours later and deposited on aluminum foil as described below. At this time the xenon activity was virtually pure Xe^{122}. Decay curves showed a straight line decay of 19 ± 0.5 hours over more than 5 half lives. The amount of 18-hour Xe^{125}

is slight because the (p,3n) reaction cross section is down at this high proton energy. Separation of iodine activity at this time showed only the 3.5-minute I^{122} activity originally reported by Marquez and Perlman[49] and studied more completely by Young, Pool and Kundu.[50] The positron spectrum of the Xe^{122}-I^{122} mixture mounted on 0.1-mil aluminum foil was studied in the double-focusing beta-ray spectrometer.

Figure 33 is the Fermi-Kurie plot of this spectrum showing a single component with an endpoint energy of 3.12 ± 0.04 Mev. The calibration of the spectrometer was checked with the 1.97-Mev positron of Cs^{130}.[26] The Xe^{122} decays by K capture and the 3.12-Mev positron group is assigned to the 3.5-minute I^{122} in equilibrium with it. Our positron energy agrees with the 3.08 ± 0.1-Mev value determined by Young, Pool and Kundu[50] who used absorption methods.

Figure 34 shows the gamma spectrum taken with the sodium iodide scintillation spectrometer. The only prominent peaks are a 182-kev gamma peak, a smaller gamma peak at 235 kev and the annihilation gamma-ray peak resulting from the positrons of I^{122}. No gamma rays of higher energy were observed. The 182- and 235-kev gamma peaks may be assigned to the decay of Xe^{122} as proved by the curves of Fig. 35. The upper curve taken on the Xe^{122}-I^{122} mixture shows these two peaks while the lower curve taken on a pure sample of I^{122} shows only the Compton smear of the Compton scattered annihilation radiation. This result is not surprising since Te^{122} is an even-even nuclide with an excited state at 568 kev[5] which in all likelihood is the first excited state, according to the systematics of the excited states of even-even nuclei as discussed by Scharff-Goldhaber.[17] The conversion electrons of the 182-kev gamma ray were observed (see below).

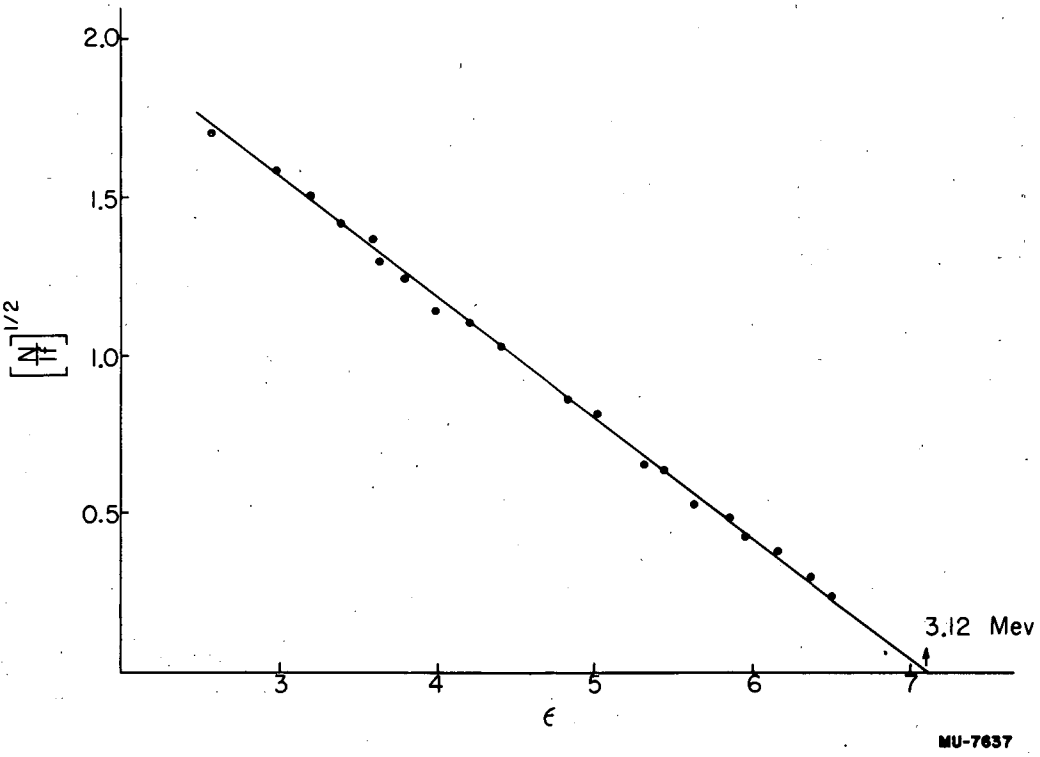

Fig. 33. Fermi-Kurie plot of positron spectrum of I^{122} obtained on Xe^{122}-I^{122} sample.

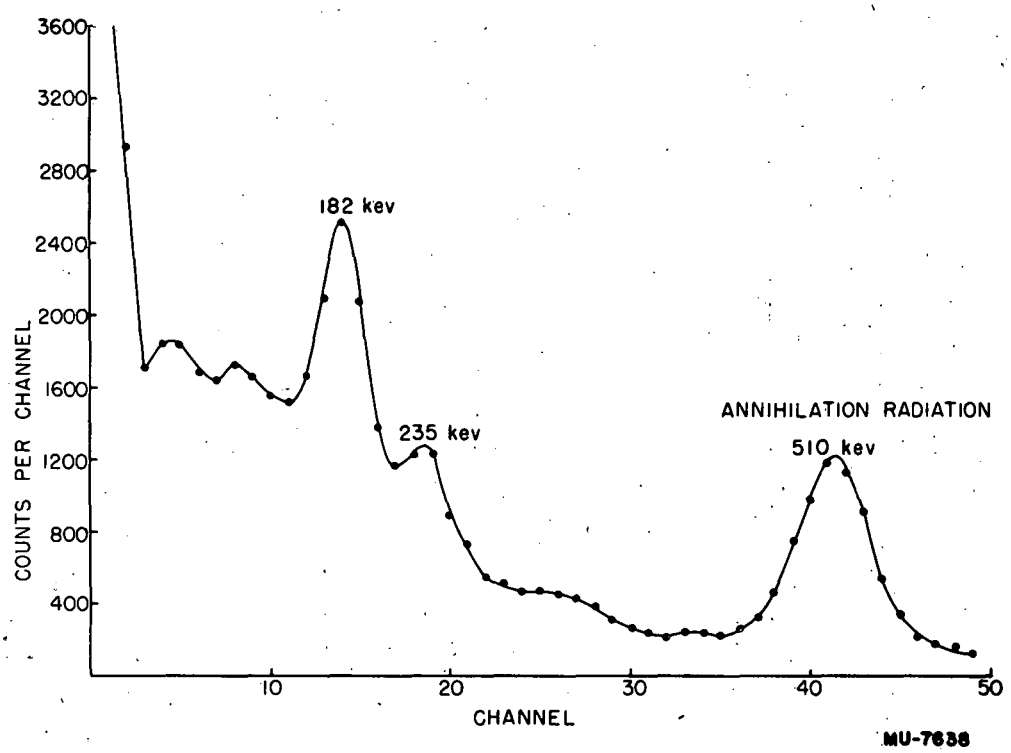

Fig. 34. Gamma spectrum of Xe^{122} taken on scintillation spectrometer.

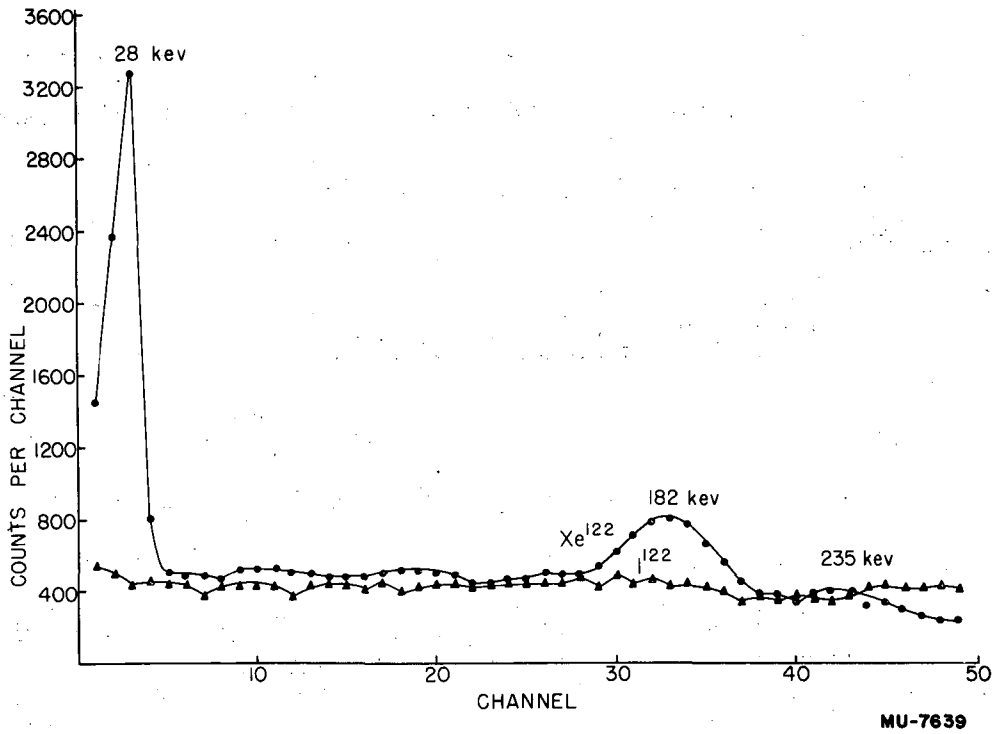

Fig. 35. Gamma spectrum of Xe^{122}-I^{122} mixture and of pure I^{122} showing assignment of 180-kev radiation to Xe^{122}. Triangular points represent pure I^{122}. The 28-kev peak is K x-radiation.

B. Xenon 123 - Iodine 123

When the xenon fraction was separated from the potassium iodide target about 4 hours after the end of the bombardment 1.8-hour Xe^{123} and 19-hour Xe^{122} accounted for the greater part of the activity. It was possible to study the radiations of Xe^{123} in such mixtures, but samples which were much purer, although less intense, could be isolated by an alternate method; namely, isolation of Xe^{123} daughter activity from the decay of Cs^{123}.

In a previous publication[51] it was shown that 6-minute Cs^{123} is produced by the bombardment of calcium iodide with 130-Mev helium ions. From cesium fractions isolated quickly immediately after bombardment it is possible to separate Xe^{123} activity >90 percent pure by radioactivity. Some 18-hour Xe^{125} is present from the decay of 45-minute Cs^{125} also produced in the bombardment.

The decay of the activity as followed in a GM tube showed the 1.8-hour decay of Xe^{123} superimposed on the growth and decay of the 13-hour I^{123} daughter.

The gamma spectrum of Xe^{123} shows a major peak of K x-rays from the electron capture decay of Xe^{123}, a gamma ray of 150-kev energy and a small peak of annihilation radiation (not shown) (see Fig. 36). Repeat runs on the gamma spectrum over a period of 24 hours showed the emergence of the 159-kev gamma peak[52] of I^{123} as the 150-kev gamma radiation of Xe^{123} decayed. The conversion electrons of these two gamma rays were also observed (see below).

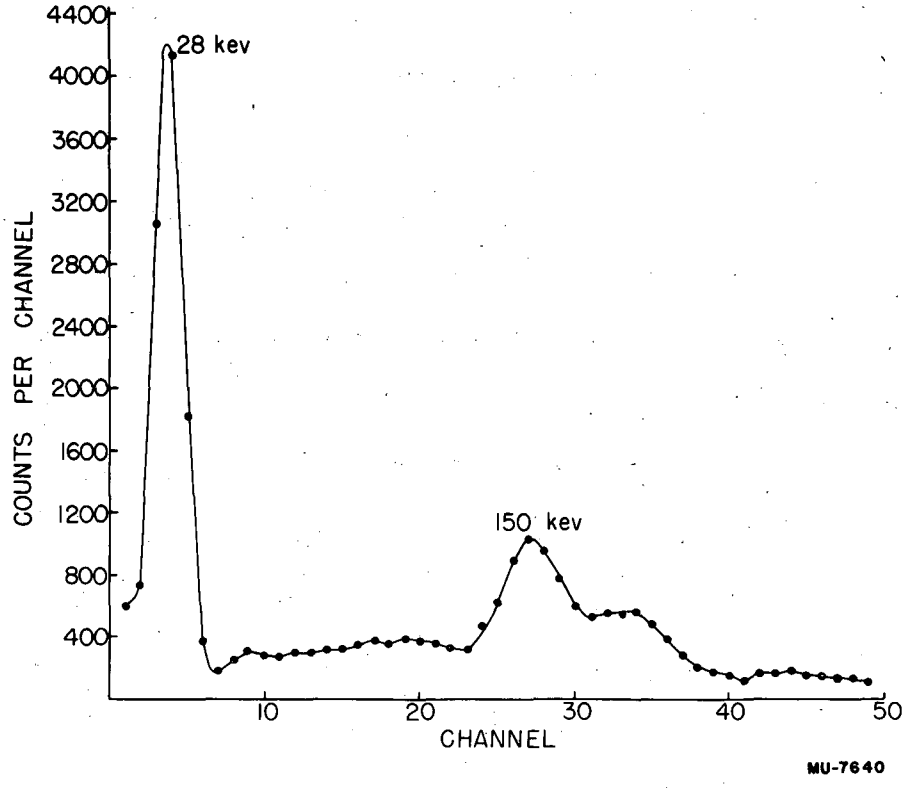

Fig. 36. Gamma spectrum of Xe^{123} sample isolated from Cs^{123}.

A beryllium absorption curve on the positron activity gave a value of 1.8 Mev as the endpoint energy of the positron. The energy of the positron as determined by the anthracene crystal spectrometer is 1.7 ± 0.1 Mev.

C. Xenon 121 and Iodine 121

Xenon samples isolated immediately after bombardment contained a high proportion of 40-minute Xe^{121}. Iodine daughter activity isolated from the xenon fraction within 1 hour of the end of bombardment contained 3.5-minute I^{122}, 1.5-hour I^{121} and 13-hour I^{123}, but after the rapid decay of the I^{122}, the principal activity was I^{121}. A gamma spectrum of such an iodine sample is shown in Fig. 37. In addition to the annihilation radiation a gamma ray of 210 kev is observed; the conversion electrons of the gamma ray were also observed as reported below. Marquez and Perlman[49] had reported conversion electrons of this gamma ray in the original report on the properties of I^{121}.

It was determined that Xe^{121} emits positrons by plotting the decay of the annihilation peak of gamma spectra determined on xenon samples isolated within 1 hour of the end of the bombardment. A 40 ± 10-minute component was resolved from this curve.

D. Conversion Electrons of Xe^{121}-Xe^{122}-Xe^{123} Mixture

The conversion electrons of several of the gamma rays mentioned above were measured in the beta-ray spectrometer using a xenon sample collected on a 1/4-mil aluminum foil 1 hour after the finish of a 1-hour bombardment of potassium iodide with 100-Mev protons. The conversion-

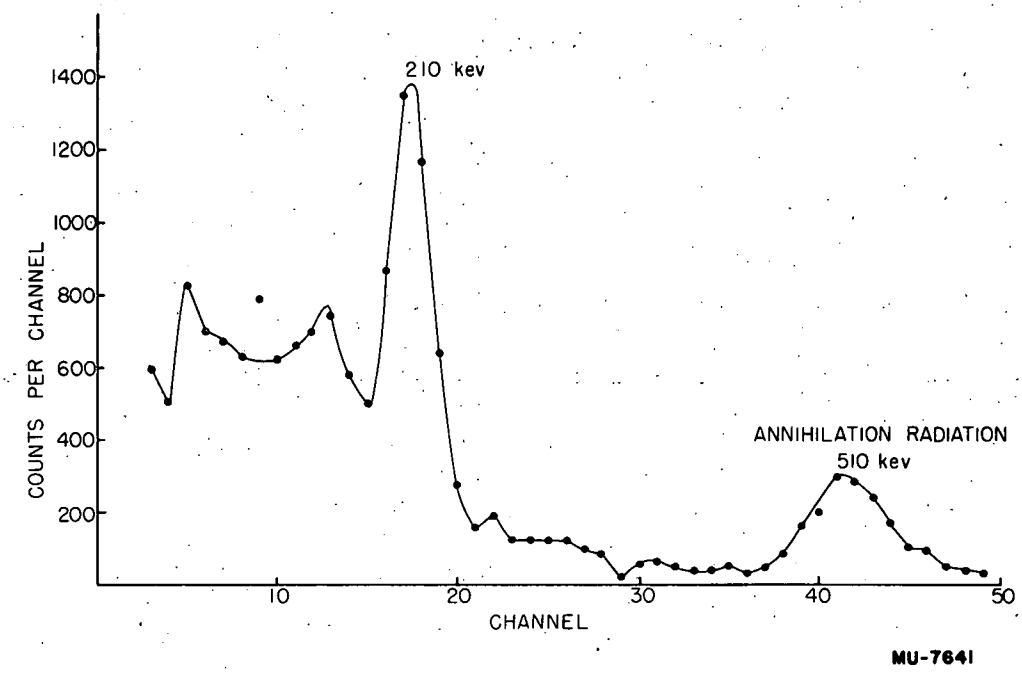

Fig. 37. Gamma spectrum of I^{121}.

electron spectrum was determined about 2 hours after the end of the bombardment (see Fig. 38) and this determination was repeated 5 times over the next 24-hour period. The numbered peaks of Fig. 38 are listed in Table 2. The observed half life and the probable assignment of the isotopes and conversion shell are listed in Table 2.

VII. EXPERIMENTAL METHODS

A. Cyclotron Targets

During the course of the work cesium isotopes were produced by $I^{127}(\alpha,xn)Cs^{<131}$ reactions by bombarding I^{127} (100 percent abundance) in the form of calcium iodide with helium ions. The calcium iodide powder was wrapped in 1-mil thick aluminum or 1/2-mil platinum foil. In the 184-inch cylotron bombardments helium ions ranging from 60 to 150 Mev were obtained by inserting the target to the correct radial setting. For studies of Cs^{130}, calcium iodide was bombarded with 20-Mev helium ions in the 60-inch cyclotron.

The neutron-deficient isotopes of xenon were produced by $I^{127}(p,xn)Xe^{>123}$ reactions by bombarding I^{127} in the form of potassium iodide with 100-Mev protons in the 184-inch cyclotron. In the studies on 34-day Xe^{127}, samples were made by the $I^{127}(p,n)Xe^{127}$ reaction by bombarding potassium iodide with 10-Mev protons in the 60-inch cyclotron or in the linear accelerator.

Molybdenum 90 activity was produced by the $Nb^{93}(p,4n)Mo^{90}$ reaction by bombarding Nb^{93} (100 percent abundance) in the form of 5-mil foil. Protons of 80-Mev energy from the 184-inch cyclotron were used for bombardment.

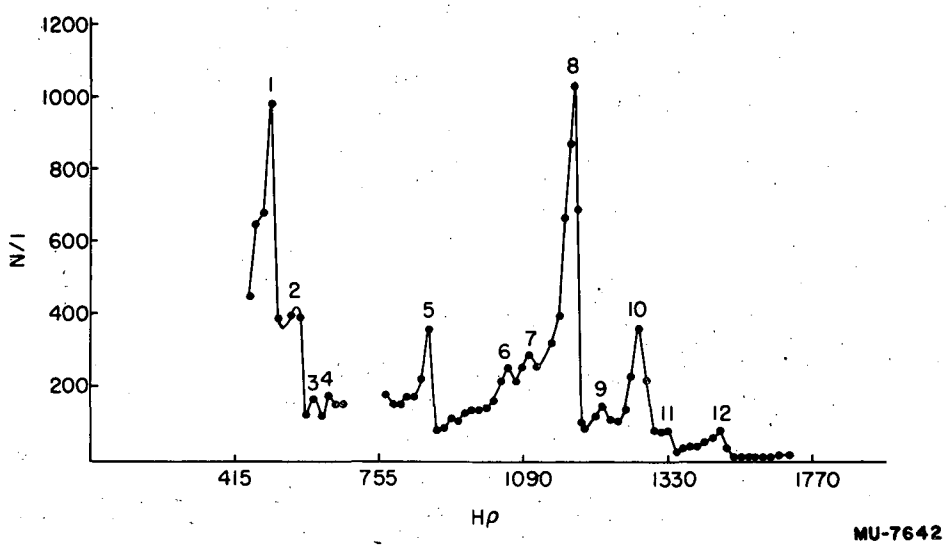

Fig. 38. Conversion electrons in Xe^{121}-Xe^{122}-Xe^{123} mixture.

Table 2

Conversion Electrons of Xe^{121}-Xe^{122}-Xe^{123} Mixture

Peak number	Energy (kev)	Approximate observed half life	Assignment of parent isotope	Conversion shell	Gamma ray energy	Gamma ray energy from scintillation spectrometer
1	22.2	4 hr	mixture	Auger electrons	-	-
2	27.0	3 hr				
3	31.0	2 hr				
4	34.5	0.5 hr	?	-	-	-
5	63.2	45 min	Xe^{121}	K	95	-
6	91	50 min		L	96	
7	97	1.8 hr	Xe^{123} (?)	-	-	-
8	115.5	2.5 hr	Xe^{123}	K	148	150
10	144	2 hr		L	147	
9	128	20* hr	I^{123}	K	160	160
11	155	14 hr	Xe^{122}	K	187	182
12	180	3 hr	I^{121}	K	212	210

*The limited number of points taken and the imperfect resolution of the electron peaks made it impossible to observe the growth of this peak from its 1.8-hour parent.

In addition to the reaction mentioned above, Mo^{90} was also produced by the bombardment of bromine with accelerated nitrogen ions in the 60-inch cyclotron which gives a beam of hextuplicately charged nitrogen[53] ions which have a continuous spread of energy with a maximum of 140 Mev. Sodium bromide wrapped in thin tantalum foil was bombarded to carry out the following reactions.

$$_{7}N^{14} + {}_{35}Br^{81} \rightarrow {}_{42}Mo^{91} + 4\,{}_{0}n^{1}$$

$$_{7}N^{14} + {}_{35}Br^{81} \rightarrow {}_{42}Mo^{90} + 5\,{}_{0}n^{1} .$$

Niobium 89 and niobium 90 activities were produced by the bombardment of 5-mil zirconium foils with protons in the Berkeley linear accelerator or by bombarding silver foils with 340-Mev protons in the 184-inch cyclotron. In addition, bombardment of niobium foils with 100-Mev protons, or yttrium oxide powder wrapped in 1-mil aluminum foils with 60-Mev helium ions also produced niobium activity by the reactions $Nb^{93}(p,pxn)Nb^{>91}$ and $Y^{89}(\alpha,xn)Nb^{>91}$, respectively.

B. Radiochemical Procedures

Cesium.--The calcium iodide target was dissolved in water and, after placing the solution in ice water, gaseous HCl was passed in until the solution was saturated. Several drops of 0.4 M silicotungstic acid were added to precipitate free silicotungstic acid. This precipitate was centrifuged from the solution; it carried the cesium activity. The silicotungstic acid was dissolved in 2 drops of water and reprecipitated

by the addition of cold saturated HCl for more purification. Finally the silicotungstic acid dissolved in 0.5 ml water was passed through a 1 cm x 4 mm column of Dowex-50 cation exchange resin. The cesium adsorbed on the resin while the silicotungstic acid passed through. The resin column was washed free of silicotungstic acid with distilled water following which the cesium activity was quickly desorbed from the column with a few drops of 6 M HCl. The cesium so obtained was carrier free. This procedure is described more fully elsewhere.[54]

Molybdenum.--For the isolation of molybdenum, bombarded niobium foils were dissolved in a mixture of concentrated HNO_3 + HF and the solution was evaporated to dryness. The residue was taken up in 6 M HCl and centrifuged to remove Nb_2O_5, and molybdenum was extracted from the clear supernatant with an equal volume of diethyl ether. The extraction was repeated thrice. Ether from the three extractions was evaporated over 5 ml of distilled water which was then saturated with HCl gas. The resulting solution was run through a 4 cm x 0.5 cm column of Dowex-1 anion-exchange resin previously equilibrated with concentrated HCl. The resin column was then washed free of niobium with 5 M HCl following which the molybdenum activity was eluted from the column with 1 M HCl.

In the case of sodium bromide targets bombarded with nitrogen ions, the target was dissolved in 6 M HCl and molybdenum was extracted with diethyl ether. Thereafter, the chemical procedure was the same as described in the preceding paragraph.

Niobium.--In the case of zirconium targets, the foils were dissolved in HF acid and all the zirconium was precipitated from the solution as

barium fluozirconate by adding Ba^{++}. Precipitation of LaF_3 from the solution by addition of La^{+3} ion removed yttrium radioactivity from the solution. The resulting solution was saturated with HCl and niobium was extracted into di-isopropyl ketone.

In order to isolate niobium activity from the spallation products of silver, the silver targets were dissolved in 10 M HNO_3 and niobium was coprecipitated on MnO_2 by adding a few drops of a dilute solution of potassium permangante and heating the solution in a water bath. The precipitate of MnO_2 was dissolved in concentrated HCl and niobium was extracted into di-isopropyl ketone. Part of the manganese was extracted by the ketone.

Alternatively, to the solution of silver in 10 M HNO_3, a few drops of $La(NO_3)_3$ were added and the solution was saturated with ammonia gas. After centrifuging the precipitate of $La(OH)_3$ was removed, washed with distilled water and dissolved in concentrated HCl. The resulting solution was run through a column of the anion-exchange resin Dowex-1, the column was washed with concentrated HCl a few times, and finally niobium was desorbed from the column with 5 M HCl.

C. Glow-Discharge Method

Samples of xenon activity produced in the decay of cesium isotopes or xenon produced directly by bombardment of iodine with protons were deposited on thin metallic foils by the method developed and used extensively by Momyer and Hyde[46] for the study of isotopes of emanation. This method makes use of a glow-discharge tube of the type shown in Fig. 39.

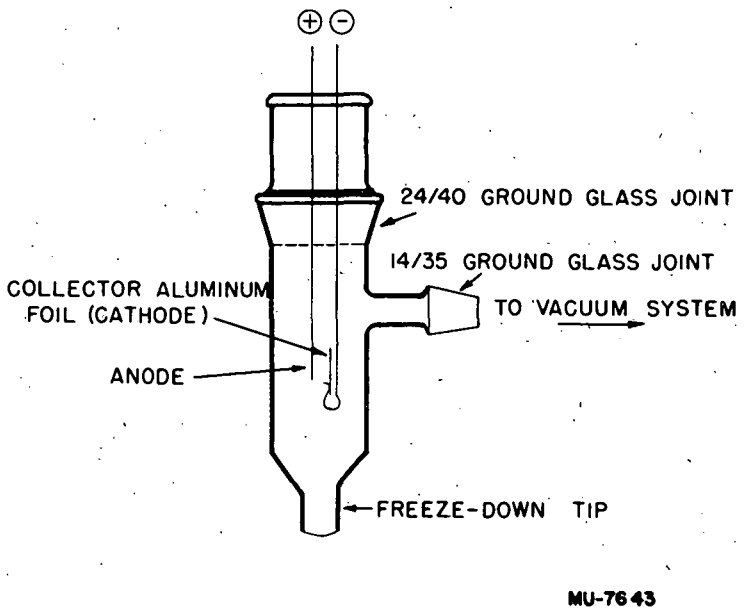

Fig. 39. Glow-discharge tube used to deposit xenon on aluminum foils.

Figure 40 shows the glass vacuum system used to isolate xenon from the cyclotron targets. The base pressure in the manifold was reduced to 10^{-5} mm mercury by a mercury diffusion pump and a Cenco Hyvac mechanical forepump. The potassium iodide target was dissolved in the closed dissolving tube A by introducing water from the dropping funnel. Because of a highly reduced pressure maintained in the manifold, the off gases were pumped through trap C cooled by a dry ice-acetone cooling bath which removed water vapor, and through traps D, E and F cooled with liquid nitrogen which condensed the xenon. After 2 or 3 minutes stopcock B was closed and the total pressure in the entire system including the glow-discharge tube was reduced to 10^{-4} to 10^{-5} mm mercury. Then that part of the system including traps D, E, F, and the discharge tube were isolated from the rest of the system by suitable manipulation of the stopcocks and the xenon activity was distilled into the discharge tube by placing a liquid nitrogen cooling bath on the freeze-down tip and by warming the traps D, E and F to room temperature. Then stopcock G was closed and the xenon and other condensed material was allowed to vaporize in the glow-discharge tube. Usually enough inert vapors were condensed during this crude fractionation that the total pressure in the tube rose to the region 200 to 1000 microns mercury. When this was not true air was bled in until the pressure rose to this value as read on a thermocouple gauge attached to the side tube of the glow-discharge tube. A dc potential of 300 to 800 volts with a limiting resistor of 50,000 ohms was placed across electrodes (see Fig. 39) to initiate and maintain a glow discharge in the tube. The xenon atoms were ionized and collected on

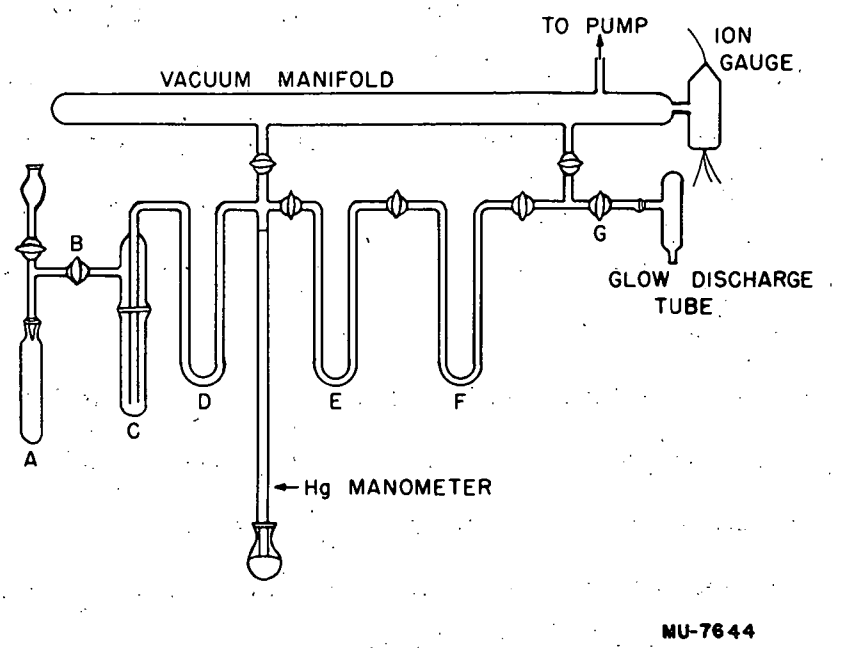

Fig. 40. Glass vacuum system used to isolate xenon activity.

aluminum foils of 0.1-mil thickness clipped to the negative electrode. A collection time of about 5 minutes served to affix a variable percentage (2 to 10 percent) of xenon activity on both sides of this foil. The excess activity was pumped back into trap F and condensed with liquid nitrogen. The glow-discharge tube was then removed from the line and opened. The xenon activity on the foil remained affixed indefinitely unless the foil was warmed above room temperature. It is believed that proper redesign of this method to allow better cooling of the electrodes during deposition would make possible quantitative collection of tracer xenon activity.

In cases where it was desired to milk xenon activity from a solution of cesium activity, the solution of the parent activity took the place of the dissolver vessel A. The xenon was then swept from the solution by running a slow stream of air through it, the rest of the procedure being the same as described above.

When it was necessary to milk iodine activities from the xenon parent, the glow-discharge tube was replaced by a glass tube sealed at one end. Xenon was collected in this tube by cooling it in a liquid nitrogen cooling bath and warming traps D, E and F to room temperature. It was then allowed to sit there for a suitable period and then was transferred to trap F cooled by a liquid nitrogen bath by opening the stopcock G and warming the tube with a dry ice-acetone cooling bath. Iodine produced by the decay of the xenon parent remained in the tube at this temperature while xenon was distilled over. The tube containing iodine activity was then removed from the line and iodine was removed by washing the sides with a dilute solution of ammonia.

D. Instruments

Time-of-flight mass spectrometer.--The mass assignment and isotopic separation reported in this report were done on a time-of-flight isotope separator in use in this laboratory. An unpublished account of the instrument has been given by Dr. W. E. Glenn[55] and a full description will be published by Drs. M. C. Michel and D. H. Templeton[56] shortly. The instrument is a medium-resolution, high-transmission time-of-flight mass separator which is used to collect samples of radioactive isotopes in the mass region 65 to 270. The overlapping of one mass on adjacent masses is less than 1 percent of peak intensity.

In the case of cesium isotopes, the ion source was a tungsten ribbon surface on which carrier-free Cs_2SO_4 was evaporated. The tungsten ribbon was heated electrically in the source region to produce thermal ions with a low spread in energy.

For the purpose of collection of active isotopes, the ions are discharged on a platinum counting plate introduced in the collection end at ground potential. The majority of ions formed are of the type M^+ which are nonvolatile when discharged and remain as a thin uniform covering on the metal surface exposed to the beam. The metal plate can thus be used directly for counting measurements.

It was thus possible to get separated single isotopes of purity greater than 99 percent with a yield of from 5 to 10 percent of the activity placed on the source filament.

It is possible to work rapidly in order to assign short half lives.

Beta-ray spectrometers.--Two precision beta-ray spectrometers were used. The first was a 25-cm radius of curvature spectrometer of the double-focusing type proposed by Svartholm and Siegbahn[57] and by Shull and Dennison.[58] A side-window GM tube was used as a detector. This tube had a 0.005-inch platinum central wire; a thin window of vinyl plastic supported on a grid of 0.001-inch tungsten wires and was filled to a regulated pressure of 8.8 cm with a gas mixture 90 percent argon and 10 percent ethylene. A more complete description of the instrument is given in an unpublished report by O'Kelley.[59]

The calibration for negative electrons was checked with the K line of the 662-kev gamma ray of Cs^{137}, and the K line of the 80.1-kev gamma ray of I^{131}. The transmission of the spectrometer under the conditions used was approximately 0.3 percent. For the study of the positrons the calibration was checked with the 1.97-Mev positron of Cs^{130}. We are indebted to Dr. Thomas O. Passell for major assistance in the use of this instrument.

Cesium samples for the spectrometer were prepared by evaporating the carrier-free activity dissolved in HCl on gold leaf of 87 $\mu gm/cm^2$ thickness. The gold leaf was supported on a brass ring.

For the study of the positrons and electrons of Cs^{127} the results on the double-focusing spectrometer were supplemented by studies carried out on a magnetic lens-type spectrometer with somewhat lower resolution but considerably higher transmission (about 1 percent). This instrument was made available to us through the kindness of the chemistry division of the California Research and Development Corporation and we are particularly indebted to Mr. James Olsen and Dr. Grover D. O'Kelley for assistance in its use. The cesium samples used in this instrument

were mounted on a single layer of tygon film (about 20 $\mu gm/cm^2$) supported by a plastic ring.

Scintillation spectrometer.--The gamma ray scintillation spectrometer used in this work was assembled by A. Ghiorso and A. E. Larsh of this laboratory. The gamma detection initially occurred in a 1.5-inch diameter by 1-inch thick crystal of sodium iodide (thallium activated) procured from Harshaw Chemical Company. The photomultiplier coupled to the crystal was a Dumont-6292 tube. The mounting of the crystal followed methods described by Borkowski.[60] On the side of the crystal facing the photomultiplier tube was affixed a quartz disk; a layer of oil between the quartz and the outside surface of the tube provided optical coupling. The other surfaces of the tube were packed into a reflecting layer of magnesium oxide. The whole assembly was mounted in an aluminum-lined lead shield on top of a standard GM counter 5-position shelf assembly. Incident gamma rays penetrated a thin foil of beryllium (about 150 mg/cm^2) and a thin layer of magnesium oxide (about 1/16 inch) before entering the crystal.

The output pulse from the photomultiplier was amplified in a pre-amplifier then in a linear amplifier. The final pulse is introduced to a 50-channel differential pulse-height analyzer. The analyzer based on a novel use of a 6BN6 as one arm of a gated univibrator is a new design of Ghiorso and Larsh. After proper alignment the channel width stability (operating at a 5-volt channel width) was better than 1 percent and it remained so for a period of weeks. Gain and bias controls permitted the inspection of any predetermined energy intervals with the full 50 channels. In order to calibrate the apparatus at any particular gain

and bias settings, use was made of the known energies in the gamma spectrum of various standards such as annihilation radiation from Na^{22}, 662-kev radiation from Cs^{137}, 60-kev radiation from Am^{241}, 184-kev radiation from U^{235}, etc. Further details of this equipment will be obtainable in a forthcoming publication of Ghiorso and Larsh.[61]

The positron energy measurements were made with a 1/4-inch thick anthracene crystal used in connection with the above equipment.

The gamma-gamma coincidence spectrometer incorporated the above equipment in combination with a second single-channel pulse-height analyzer. The sample was mounted between two sodium iodide photomultiplier tube detectors. Pulses resulting from events in the gate crystal were fed to the single-channel analyzer and those corresponding to a selected gamma energy interval were used to gate a coincidence circuit. Pulses arriving from the second crystal in coincidence with these were fed to the 50-channel analyzer. Hence the gamma spectrum in coincidence with a particular gamma ray could be determined quickly.

VIII. ACKNOWLEDGMENTS

The author takes this opportunity of thanking Dr. E. K. Hyde for his guidance, encouragement and assistance throughout the course of these experiments. He also wishes to express his gratefulness to Professor G. T. Seaborg for his general supervision and guidance. The indispensable cooperation of Dr. M. C. Michel in the mass separations, Dr. T. O. Passell for beta-ray spectroscopy, Dr. F. Asaro and Mr. F. S. Stephens for gamma-gamma coincidence studies is also acknowledged.

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He also wishes to express thanks to James T. Vale and Lloyd Hauser and members of the 184-inch cyclotron crew for the numerous 184-inch cyclotron bombardments. Similarly, acknowledgment is made to G. B. Rossi, W. B. Jones, A. Ghiorso, and members of the 60-inch cyclotron crew for the nitrogen ion and carbon ion bombardments. The linear accelerator bombardments were carried out through the courtesy of W. W. Olson, R. D. Watt and the accelerator crew.

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IX. REFERENCES

1. O. Hahn, Chem. Berichte 54, 1131 (1921).
2. B. V. Kurchatov, I. Kurchatov, L. Myssowski, and L. Roussinow, Compt. rend. 200, 1201 (1935).
3. E. Amaldi, O. D'Agostino, E. Fermi, B. Pontecorvo and E. Segrè, Ricerca Scientifica 61, 581 (1935).
4. M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).
5. M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179 (1952).
6. C. F. von Weizsäcker, Naturwissenschaften 24, 813 (1936).
7. P. Axel and S. M. Dancoff, Phys. Rev. 76, 892 (1949).
8. V. F. Weisskopf and J. M. Blatt, Theoretical Nuclear Physics (John Wiley and Sons, New York, 1952).
9. V. F. Weisskopf, Phys. Rev. 83, 1073 (1951).
10. R. Montalbetti, Can. J. Phys. 30, 660 (1952).
11. Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. 83, 79 (1951).
12. M. G. Mayer, Phys. Rev. 78, 16, 22 (1950).
13. Haxel, Jensen, and Suess, Z. Physik 128, 301 (1950).
14. O. Kurath, Phys. Rev. 80, 98 (1950).
15. I. Talmi, Phys. Rev. 82, 101 (1950).
16. L. W. Nordheim, Revs. Modern Phys. 23, 315 (1951).
17. G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).
18. I. Bergström, Arkiv för Fysik 5, 191 (1952).
19. R. W. Fink, F. L. Reynolds, and D. H. Templeton, Phys. Rev. 77, 614 (1950).
20. A. H. Wapstra, N. F. Verster, and M. Boelhouwer, Physica XIX, 138 (1953).

21. M. C. Michel and D. H. Templeton, Phys. Rev. 93, 1422 (1954).

22. A. H. Wapstra, Physica 19, 671 (1953).

23. Creutz, Delasasso, Sutton, White, and Barkas, Phys. Rev. 58, 481 (1940).

24. I. Bergström, Phys. Rev. 82, 111 (1951).

25. J. R. Risser and R. N. Smith, private communication from K. Lark-Horowitz (1948) as reported by Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).

26. Smith, Mitchell, and Caird, Phys. Rev. 87, 454 (1952).

27. E. K. Hyde and C. A. Levine, unpublished data (1951).

28. P. K. Kofstad, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-2265 (June 30, 1953)(unpublished).

29. Shore, Bendel, Brown, and Becker, Phys. Rev. 91, 1203 (1953).

30. H. G. Hicks and R. S. Gilbert, California Research and Development Corporation Unpublished Reports CRD-R-57 and MTA-33 (April 1953).

31. G. E. Boyd and B. H. Ketelle, unpublished data (1951) as reported by Hollander, Seaborg, and Perlman, Revs. Modern Phys. 25, 469 (1953).

32. Miller, Hamilton, Putnam, Haymond, and Rossi, Phys. Rev. 80, 486 (1950).

33. R. M. Diamond, Phys. Rev. 95, 410 (1954).

34. S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

35. A. de-Shalit and M. Goldhaber, Phys. Rev. 92, 1211 (1953).

36. R. M. Diamond, Phys. Rev. 89, 1149 (1953).

37. B. Kahn and W. S. Lyon, Nucleonics 11, 61 (1953).

38. G. E. Boyd, Oak Ridge National Laboratory Report ORNL-229 (February 1949)(unpublished) as reported by Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).

39. D. H. Kundu and M. L. Pool, Phys. Rev. 76, 183 (1949).

40. L. Jacobson and R. Overstreet, Radiochemical Series: The Fission Products (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Engineering Series, Plutonium Project Record, Vol. 9, Div. IV, Paper No. 91.

41. Huffman, Iddings, and Lilly, J. Am. Chem. Soc. 73, 4474 (1951).

42. K. A. Kraus and G. E. Moore, J. Am. Chem. Soc. 73, 9 (1951).

43. Hicks, Stevenson, and Gilbert, private communication.

44. Beling, Newton, and Rose, Phys. Rev. 86, 797 (1952); 87, 1144 (1952) Errata.

45. Broyles, Thomas, and Haynes, Phys. Rev. 89, 715 (1953).

46. F. F. Momyer, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-2060 (February 1953) (unpublished).

47. D. E. Tilley, Abstract 77, June Meeting, Roy. Soc. Canada (1952).

48. B. Dropesky and E. O. Wiig, Phys. Rev. 88, 683 (1952).

49. L. Marquez and I. Perlman, Phys. Rev. 78, 189 (1950).

50. Young, Pool, and Kundu, Phys. Rev. 83, 1060 (1951).

51. H. B. Mathur and E. K. Hyde, Phys. Rev. 95, 708 (1954).

52. Mitchell, Mei, Maienschein, and Peacock, Phys. Rev. 76, 1450 (1950).

53. Rossi, Jones, Hollander, and Hamilton, Phys. Rev. 93, 256 (1954).

54. E. K. Hyde, J. Am. Chem. Soc. 74, 4181 (1952).

55. W. E. Glenn, Ph.D. Thesis, University of California Radiation Laboratory Report UCRL-1628 (January 1952).

56. M. C. Michel and D. H. Templeton, to be published.

57. N. Svartholm and K. Siegbahn, Arkiv. Mat. Astron. Fysik 33A, No. 21 (1946); see also Hedgran, Siegbahn, and Svartholm, Proc. Phys. Soc. (London) 63A, 960 (1950).

58. F. Shull and D. Dennison, Phys. Rev. 71, 681 (1947); 72, 256 (1947).

59. G. D. O'Kelley, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-1243 (March 1951)(unpublished).

60. C. J. Borkowski, Oak Ridge National Laboratory Unpublished Report ORNL-1336 (September 1952).

61. A. Ghiorso and A. E. Larsh, Jr., to be published.