Studies on the Isomeric Pair, Nb^{89m} and Nb^{89}*

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By bombardment of zirconium, niobium, yttrium, and silver targets in the 184-inch cyclotron it has been possible to produce the isomer pair Nb^{89m} and Nb^{89}. The isomerism arises from the odd 41st proton below the 50-proton closed shell of the single-particle nuclear model. $Nb^{89}(g_{9/2})$ is a 1.9-hour activity emitting 2.85-Mev positrons to produce Zr^{89}. The yield of the $Nb^{89m}(p_{\frac{1}{2}})$ is about 30–100 fold less and its radiations were not directly observed. By scintillation spectrometer measurements on zirconium daughter activity it is shown that Nb^{89m} decays with about a 2-hour half-life to 4.4-minute $Zr^{89m}(p_{\frac{1}{2}})$. The $M4$ gamma transition between the two niobium levels is not observed, indicating that the level spacing is small and the upper level deactivates primarily by positron emission. The log ft values are larger than expected and are discussed in terms of the even-even core rearrangement considerations of A. De-Shalit and M. Goldhaber [Phys. Rev. **92**, 1211 (1953)]. Nb^{89} was also prepared by bombardment of NaBr with accelerated carbon ions in the 60-inch cyclotron.

I. INTRODUCTION

STUDY of the radioactivity present in the niobium fraction isolated from targets of several elements bombarded in the 184-inch cyclotron has resulted in the determination of the properties of the previously unreported isomeric pair Nb^{89} and Nb^{89m}. This information is summarized in Fig. 1. The reactions by which Nb^{89} was made and the evidence for the mass assignment are briefly given in Sec. II. Following this, a report is given of the detailed study of Nb^{89m} and Nb^{89} by means of scintillation spectroscopy. The suggested decay scheme of Fig. 5 is discussed at the end of the paper.

Independent work on these isomers has recently been prepared for publication by Diamond.[1] Our findings are similar, with one exception which is discussed later.

II. ORIENTATION EXPERIMENTS

Samples of zirconium metal were bombarded with 40-Mev protons for 30 minutes. G-M decay curves on the niobium fraction isolated from these targets within an hour of the end of the bombardment indicated that >80 percent of the activity originated with one unknown activity 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 the 1.9-hour activity to consist of positrons 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 timed milkings of the zirconium daughter activity showed that the Nb^{89} half-life was about 2–4 hours and hence Nb^{89} was identified with the 1.9-hour, ~3-Mev positron acitivity. Chemical difficulties in obtaining quantitative separation of the zirconium without appreciable loss of niobium made the results less exact than was desired.

Very similar results were obtained by studying the niobium fraction isolated from niobium foils bombarded with 90-Mev protons, or from silver foils bombarded with 340-Mev protons,[2] or from yttrium oxide targets bombarded with 60–100-Mev helium ions. In every case the two chief activities were 14.6-hour Nb^{90} and 1.9-hour Nb^{89}. The ratio by activity of Nb^{89} to Nb^{90} was roughly the same (~2–6) in each case.[3]

III. GAMMA-RAY STUDIES ESTABLISHING THE ISOMERISM

Nb^{89} has 41 protons, and according to the single-particle shell model may well exist in the states $g_{9/2}$ and $p_{1/2}$, differing in spin by 4. Hence there is the strong possibility of isomerism in Nb^{89}. The systematics of the $g_{9/2}-p_{1/2}$ separation in this region as given by Fig. 75

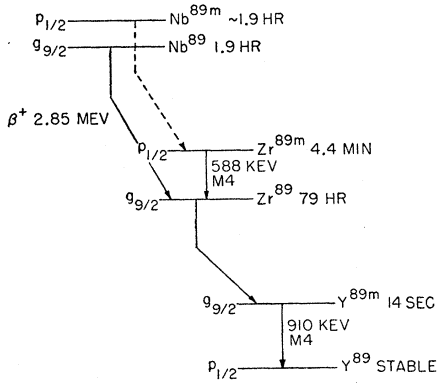

FIG. 1. Decay scheme of Nb^{89} and Nb^{89m}.

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[1] R. M. Diamond, Phys. Rev. **95**, 410 (1954).

[2] P. K. Kofstad, Ph.D. thesis, University of California Radiation Laboratory Unclassified Report UCRL-2265, June 30, 1953 (unpublished).

[3] In unpublished University of California Radiation Laboratory reports, we had indicated that we had evidence for Nb^{87} as well as Nb^{89}. On the basis of more complete study we conclude that we have no reliable evidence on the properties of Nb^{87}.

of an article on nuclear isomerism by Goldhaber and Hill[4] 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 would expect the upper state of Nb^{89} to deactivate predominantly by positron emission rather than by gamma emission. Zr^{89} is known[5] to exist in two isomeric forms by virtue of its 49 protons which give it an odd proton immediately below the 50-proton shell. The upper state is the $p_{1/2}$ state and has a half-life of 4.4 minutes for its predominant decay by emission of 588-kev gamma rays to the ground state (see Fig. 1). This ground state with spin $p_{1/2}$ decays with a half-life of 79 hours to 13-second Y^{89m}.[6] A pair of isomers occurs again in Y^{89} because of the odd proton below the 50-proton shell. $Y^{89m}(g_{9/2})$ decays by emission of a 910-kev gamma ray to stable $Y^{89}(p_{1/2})$.

From 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 taken again for 1 minute. Figure 2 shows the series of curves thus obtained. The 588-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.

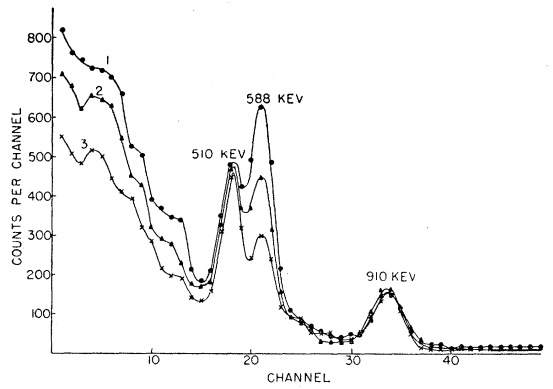

FIG. 2. Gamma spectrum of Zr^{89m} and Zr^{89} isolated from Nb^{89} after an 8-minute growth period. Curve one is a one-minute run started 8.2 minutes after an 8-minute growth period. The 588-kev gamma ray of Zr^{89m} is plainly visible. Succeeding one-minute runs show the rapid decay of this 4.4-minute activity.

After complete decay of Zr^{89m}, the gamma spectrum obtained corresponded to the annihilation radiation and the 910-kev gamma radiation expected of a sample of Zr^{89} (see Fig. 3). 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.[5] 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 $1.2\pm0.5 \times 10^{-2}$, 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 $Nb^{89}(g_{9/2})$.

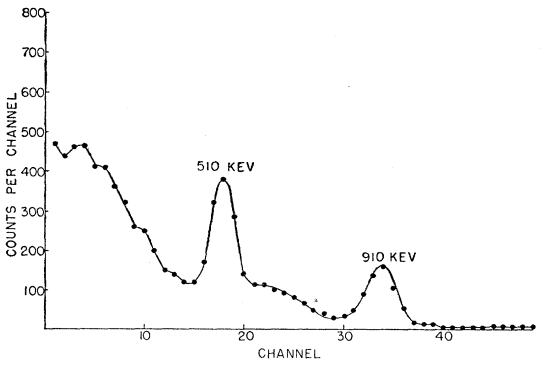

FIG. 3. Gamma spectrum of Zr^{89} daughter activity isolated from initially pure niobium fraction.

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.

[4] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

[5] Shore, Bendel, Brown, and Becker, Phys. Rev. **91**, 1203 (1953).

[6] M. Goldhaber *et al.*, Phys. Rev. **83**, 661 (1951).

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. 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 $10M$ HNO_3. Niobium was coprecipitated on MnO_2. This precipitate was dissolved in $12M$ HCl and contacted with diisopropyl ketone to extract the niobium.[7] After niobium was backextracted into water and evaporated to dryness it was taken up in a mixture of $10M$ 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. However, a significant difference 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 $(2.6\pm0.5\times10^{-2}$ 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 $\frac{3}{4}$ 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.[8] The 588-kev gamma ray was not intense enough in comparison to 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 a separate paper.

The positron end-point energy was determined by using an anthracene crystal photomultiplier combination coupled to a 50-channel pulse analyzer. Figure 4 shows a typical end-point 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.

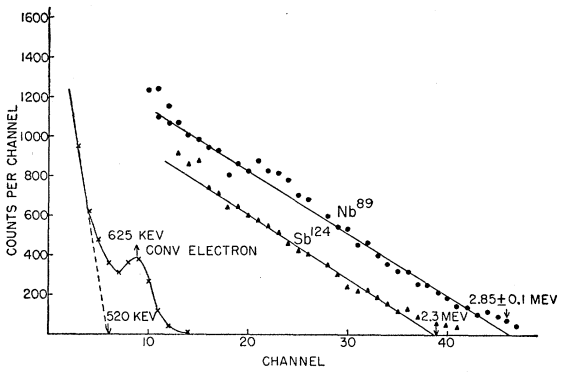

FIG. 4. Positron endpoint of Nb^{89} as determined on anthracene crystal spectrometer. Standards used were Cs^{137} and Sb^{124}.

IV. CARBON ION BOMBARDMENTS

In addition to the bombardments already mentioned, 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 by using a beam of hextuply charged carbon ions (C^{6+}).[9] 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:

$$_6C^{12}+{}_{35}Br^{81}\rightarrow{}_{41}Nb^{90}+3\ {}_0n^1,$$
$$_6C^{12}+{}_{35}Br^{81}\rightarrow{}_{41}Nb^{89}+4\ {}_0n^1.$$

After bombardment the sodium bromide was dissolved in $10M$ HNO_3 and extracted with CCl_4 to remove bromine. MnO_2 was precipitated to remove niobium. The MnO_2 was dissolved in $12M$ HCl and from this solution niobium was extracted with diisopropyl ketone. Backextraction of the niobium into water completed the purification. Resolution of a G-M decay curve extrapolated back to the end of the bombardment showed $\frac{5}{6}$ of the activity to be 1.9-hour Nb^{89} and $\frac{1}{6}$ 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}.

V. DISCUSSION

The experimental data are best summarized by the decay scheme of Fig. 1. This scheme is incomplete because the energy separation of the Nb^{89} states and the identity of the higher state have not been deter-

[7] H. G. Hicks and R. S. Gilbert, California Research and Development Corporation Reports CRD-R-57 and MTA-33, April 1953 (unpublished).

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

[9] Miller, Hamilton, Putman, Haymond, and Rossi, Phys. Rev. **80**, 486 (1950).

mined. 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,[4] it is expected that this separation in Nb^{89} is quite small and that 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 0–200-kev range with a precision spectrometer, it might be possible to learn this energy separation.

Figure 1 differs from a very similar scheme presented by Diamond[1] only in that he assigns a half-life of 0.8 hour to the $p_{1/2}$ level of Nb^{89}. This was done on 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-kev and 910-kev gamma peaks we feel they are somewhat more trustworthy.

As already mentioned, 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 0.8 hour reported by Diamond for $Nb^{89}(p_{1/2})$. It is not clear why the two methods give different results.

From Moszkowski's[10] graphs the logft 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

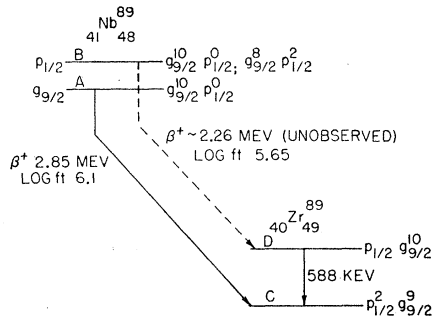

FIG. 5. Decay scheme for Nb^{89}, showing the main configurations of the protons (left) and the neutrons (right). The $g_{9/2}$ proton in A stabilizes the even-even core in a $(g_{9/2})^{10}$ configuration. The transition AC is delayed because of the necessity for rearrangement of the neutron core to produce two $p_{1/2}$ neutrons in C. B is indicated with a mixed neutron configuration, but the $(g_{9/2})^8(p_{1/2})^2$ configuration probably predominates. Hence the transition BD would be delayed.

[10] S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).

(5.0 ± 0.3) for an allowed transition. If the unknown $Nb^{89m}-Nb^{89}$ separation is neglected, the logft 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.[11] For example, the well-established $Zr^{89}(g_{9/2})\rightarrow Y^{89m}(g_{9/2})$ transition has a logft value of 6.1 and similarly, the transition $Zr^{89m}(p_{1/2})\rightarrow Y^{89}(p_{1/2})$ has a logft value of 6.85. These and other examples are discussed by De-Shalit and Goldhaber[11] who give an explanation in terms of a rearrangement of nucleons in the even-even core of an odd-A nucleus.

From their development the decay scheme of Fig. 1 can be redrawn in the form of Fig. 5.

VI. SPECTROMETER EQUIPMENT

Scintillation spectrometer.—The gamma scintillation spectrometer used during the course of this work was assembled by A. Ghiorso and A. E. Larsh of this laboratory. A sodium iodide crystal of 1.5-inch diameter and 1.0-inch thickness was used for the initial detection of the gamma ray. The photomultiplier coupled to the crystal was a Dumont-6292 tube, and mounting of the crystal was done by the methods of Borkowski.[12] The crystal-photomultiplier assembly was enclosed in a 2-inch thick lead shielding.

The output from the photomultiplier was amplified in a preamplifier and then in a linear amplifier. The final pulse was then analyzed by a 50-channel differential pulse-height analyzer designed by Ghiorso and Larsh. Calibration of the instrument was carried out with known radiations such as those from Na^{22}, Cd^{109}, Cs^{137}, U^{235}, and Am^{241} at identical gain and bias settings. A detailed description of the instrument will be given in a forthcoming publication of Ghiorso and Larsh.[13]

The positron energy measurements were made with a $\frac{1}{4}$-inch thick crystal of anthracene used in connection with the above equipment.

VII. ACKNOWLEDGMENTS

The cyclotron bombardments were carried out by James T. Vale, Lloyd B. Houser, and the 184-inch cyclotron crew except for the nitrogen beam bombardments for which we owe thanks to G. Bernard Rossi, A. Ghiorso, and the crew of the 60-inch cyclotron. The bombardments on the linear accelerator were carried out with the help of Robert D. Watt and Wendell W. Olson. We wish also to acknowledge helpful discussions with Dr. Richard M. Diamond.

[11] A. De-Shalit and M. Goldhaber, Phys. Rev. **92**, 1211 (1953).
[12] C. J. Borkowski, Oak Ridge National Laboratory Report ORNL-1336, September 1952 (unpublished).
[13] A. Ghiorso and A. E. Larsh (to be published).