Isomerism in the $N=81$ isotope ^{147}Dy†

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With the use of a helium-gas-jet capillary transport system a new (59 ± 3)-sec activity was observed in ^{14}N bombardments of ^{141}Pr. The activity was found to be associated with two coincident γ rays, 72.0 ± 0.1 and 678.7 ± 0.2 keV. The 678.7-keV transition was also observed to be in coincidence with dysprosium K x rays and its yield as a function of bombarding energy varied in the same manner as γ rays known to follow the decay of 1.9-min ^{147}Tb. From these results we conclude that the new activity is ^{147}Dy^m and that the 72.0- and 678.7-keV γ rays represent transitions in ^{147}Dy. Further, based on the systematic behavior of low-lying levels in $N=81$ odd-A nuclei, we propose that our data establish the $h_{11/2}$ and $d_{3/2}$ neutron states in ^{147}Dy to be 750.7 and 72.0 keV, respectively, above the $s_{1/2}$ ground state.

[RADIOACTIVITY ^{147}Dy^m, ^{145}Gd^m; measured $T_{1/2}$, E_γ, I_γ, $\gamma\gamma$ coin. Observed isotope ^{147}Dy. ^{147}Dy, ^{145}Gd; deduced levels, J^π.]

I. INTRODUCTION

The investigation of nuclei near the $N=82$ closed shell is of interest since it should be possible to describe their structure in terms of a single-particle framework (see, e.g., Ref. 1). Nuclei in this region with $Z \leq 63$ have been studied in several ways, including direct nuclear reactions. This particular method, however, cannot be applied to nuclei whose atomic numbers are greater than 63 because stable isotopes with $N \sim 82$ are unavailable beyond ^{144}Sm. Of necessity they must be investigated either by the in-beam γ-ray technique or else through radioactive decay.

We have recently used ^{12}C^{4+} ions from the Oak Ridge isochronous cyclotron (ORIC) to produce neutron-deficient terbium[2] and dysprosium[3,4] nuclides and have studied their decay to levels in their gadolinium and terbium daughters, respectively. One aim in Ref. 4 had been the discovery of ^{147}Dy with two purposes in mind: (1) to extend beyond ^{145}Gd the $M4$ isomeric series in $N=81$ odd-A nuclei, and (2) to study levels in the $N=82$ nucleus, ^{147}Tb. The ^{12}C^{4+} incident energy proved insufficient to reach the maximum of the ^{142}Nd(^{12}C, $7n$) excitation function. The search was therefore continued in ^{14}N + ^{141}Pr bombardments, utilizing the more energetic but less intense ^{14}N^{5+} beam. A new activity was found and assigned to ^{147}Dy^m. This information has been communicated briefly in a letter.[5] Herein, the ^{14}N + ^{141}Pr results are described and discussed in more detail.

II. EXPERIMENTAL METHOD

The apparatus consisted of a helium-gas-jet transport system.[6] A 10-m teflon capillary, i.d. of 1.3 mm, was used to extract product recoil nuclei from the gas-jet reaction chamber to a shielded area where γ- and x-ray measurements could be made.

Initial experiments were made using a collection chamber which has been described in Ref. 6. The target consisted of a 300-μg/cm^2 layer of Pr_2O_3 electrodeposited onto a 25-μm beryllium foil. First, γ-ray yields were determined as a function of bombarding energy. A Ge(Li) detector, located outside the chamber, was placed directly in front of the spot where the radioactive products were deposited by the capillary. Measurements at each bombarding energy were made continuously for a given period of time as the activity was being collected. Next, preliminary half-life determinations were made at the bombarding energy where $A=147$ nuclides reached their maximum yield, i.e., ~142 MeV. Collection cycles were varied in duration and were interrupted when the half-life measurements were in progress.

All subsequent experiments were made at the above determined ^{14}N bombarding energy. A selfsupporting 2-mg/cm^2 praseodymium foil served as the target. A different collection assembly was used, the activity being deposited onto an aluminized Mylar tape. The tape entered and exited a collection chamber through openings equipped with rubber seals. This arrangement allowed the cyclotron beam to be used continuously, since a fresh source could be collected while the previous one was being assayed. Singles and coincidence measurements were made simultaneously with two large-volume Ge(Li) detectors. Coincidence data were accumulated in a three-word γ-γ-τ list mode using the ORIC analog-to digital converter (ADC) data acquisition system. The ADC system has approximately a 9000-channel resolution capacity. It is interfaced to an in-house computer which has several discs, each with a million-word capacity. From these discs the list data were transferred to magnetic tapes for storage and subsequent analysis. Singles spectra from one of the Ge(Li) detectors were accumulated in a smaller data acquisition system in a spectrum multiscale mode for half-life information. This same acquisition system was also used for singles measurements with a Ge(Li) x-ray detector to investigate the photon energy below ~100 keV.

III. RESULTS AND DISCUSSION

In the preliminary set of experiments a new activity was observed, associated with a rather weak γ ray. Subsequently, the γ-ray energy and half-life were determined to be 678.7±0.2 keV and 59±3 sec. Figure 1 shows a portion of a spectrum measured at a ^{14}N incident energy of ~142 MeV. Data were taken in a multiscale mode, and Fig. 1 represents the sum of the first four spectra, each 15 sec in duration, measured after the end of bombardment. One can note the low intensity of the 678.7-keV peak with respect to γ rays which belong to the neighboring nuclides; ^{148}Dy, ^{148}Tb, and ^{145}Gd^m. Figure 2 shows its yield as a function of bombarding energy compared to those of γ rays that are known[2,4] to encompass most of the decay strengths of ^{146}Tb, ^{147}Tb, ^{148}Tb, and ^{148}Dy. The 678.7-keV yield curve is seen to resemble closely that of the 1397.7-keV γ ray, which represents[2] ~85% of the total decay of 1.9-min ^{147}Tb. Contrastingly, its shape differs markedly from those of $A=146$ and $A=148$ nuclides. The indication, therefore, is that the new activity has a probable mass of 147.

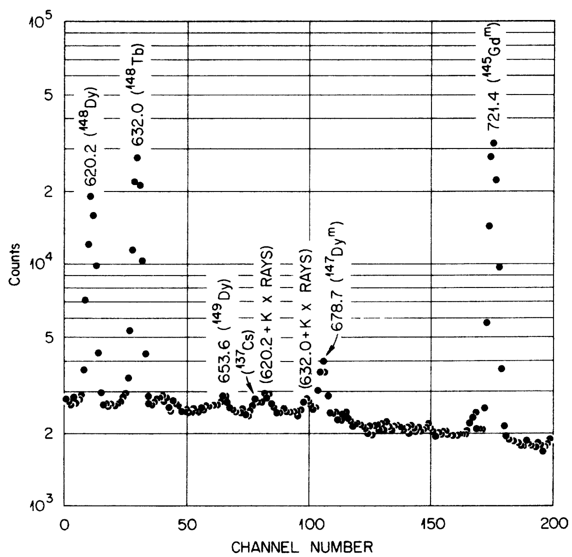

FIG. 1. Portion of a γ-ray spectrum measured for 60 sec after the end of bombardment of a ^{141}Pr target with 142-MeV ^{14}N ions. A new 678.7-keV γ ray was found to decay with a 59-sec half-life and assigned to the previously unknown activity ^{147}Dy^m.

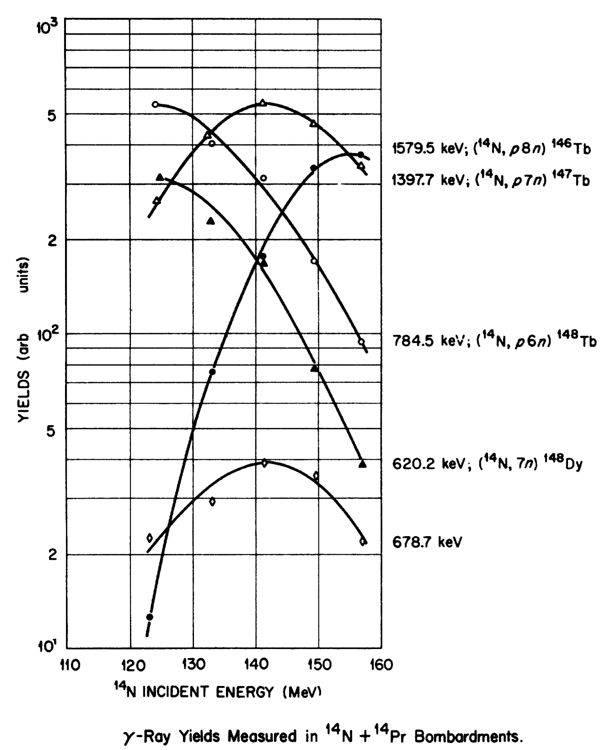

FIG. 2. Yields as a function of bombardment measured for the new 678.7-keV γ ray and for γ rays which are known to encompass most of the decay strengths of ^{146,147,148}Tb and ^{148}Dy. It is seen that the 678.7-keV yield curve resembles the one determined for the 1397.7-keV ^{147}Tb γ ray. The indication is that the new activity has a mass number of 147. Yields are not corrected for differences in half-lives; and, therefore, as discussed in the text, shorter-lived activities, e.g. ^{146}Tb, are emphasized.

The data shown in Fig. 2 are corrected for differences in beam intensity and photon efficiency but not for differences in half-life. As a result, shorter-lived activities are emphasized in yield since bombardment times were ~10 min. Yields determined from the multiscale half-life measurements will be discussed later.

The coincidence spectrum gated by the 678.7-keV transition was found to contain only K x rays and a (72.0 ± 0.1)-keV γ ray. This is shown in Fig. 3(a), which displays the low-energy portion of the coincident spectrum. The same energy ranges of spectra in coincidence with the 620.2-keV ^{148}Dy and 632.0-keV ^{148}Tb transitions (see Fig. 1) are shown in Figs. 3(b) and 3(c). In these instances only K x rays are seen, which should necessarily correspond to terbium and gadolinium x rays, respectively. The coincidence data were accumulated so that each ADC channel represented ~0.25 keV in energy. This means that K x rays of neighboring rare earth nuclides should be separated by about six channels. A systematic shift of approximately that magnitude can indeed be seen for the three spectra shown in Fig. 3 (for convenient display channel numbers have been summed by twos). The conclusion then is that the 678.7-keV γ ray is in coincidence with dysprosium K x rays. Because holmium nuclides could not be produced in the $^{14}N + ^{141}Pr$ irradiation, the new γ ray must be due to the decay of a dysprosium radioactivity.

Figure 4 shows a singles spectrum, once again representing the first minute of counting after the end of bombardment, measured with a Ge(Li) x-ray detector. The spectrum is seen to be dominated by terbium and gadolinium K x rays. Nevertheless, a distinct 46.0-keV peak is observed, an energy

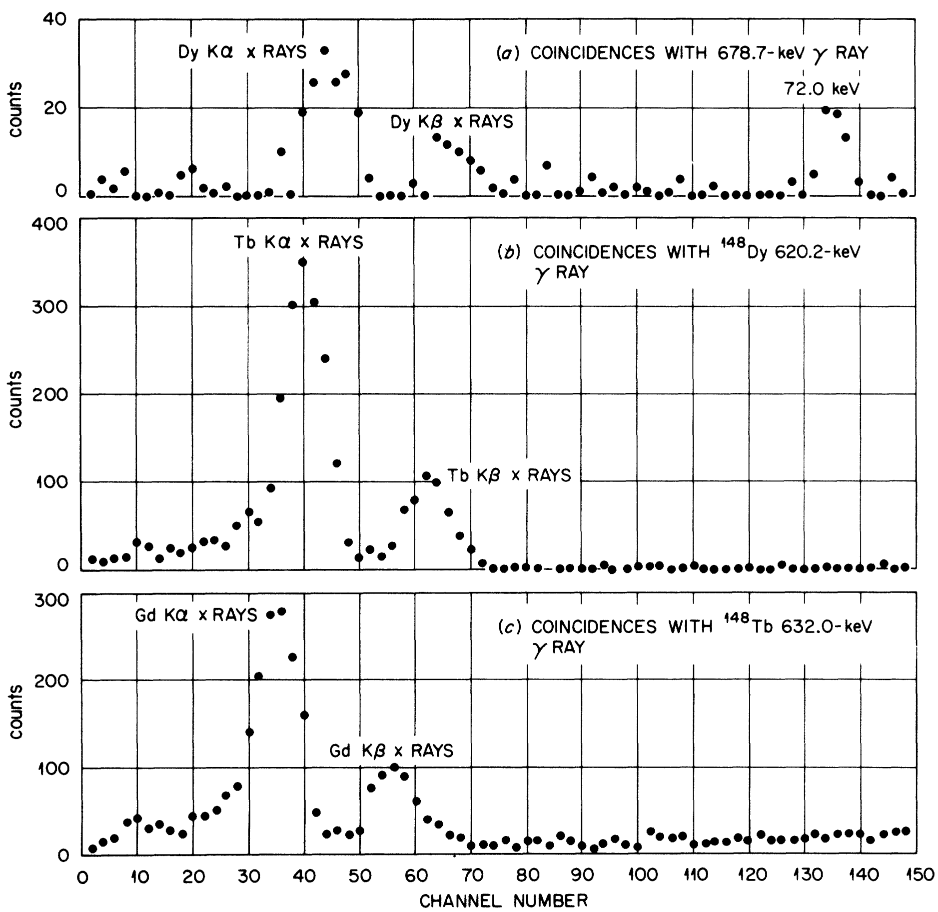

FIG. 3. Low-energy portions of coincident spectra measured in experiments involving $^{14}N + ^{141}Pr$ bombardments. Parts (b) and (c) are spectra in coincidence with γ rays known to follow the decay of ^{148}Dy and ^{148}Tb so that the observed K x rays are terbium and gadolinium x rays, respectively. Part (a) is the spectrum gated by the new 678.7-keV γ ray; here a 72.0-keV γ ray is seen together with K x rays whose energies correspond to dysprosium x rays. This can be seen by comparing the three sets of spectra and noting the systematic shift in the x-ray energies.

that corresponds to dysprosium $K\alpha_1$ x rays; smaller peaks are seen at 45.2 and 53.5 keV, matching dysprosium $K\alpha_2$ and $K\beta_2$ x ray energies. The 72.0-keV γ ray seen in Fig. 3(a) can also be observed in Fig. 4. Its half-life and that of the dysprosium $K\alpha_1$ x-ray peak were found to be ~59 sec.

Taking into account the singles and coincidence spectral measurements and yield-curve determinations we conclude that: (1) the new activity is ^{147}Dy^m, and (2) the 72.0- and 678.7-keV γ rays represent transitions in ^{147}Dy.

A 27.3-keV γ ray can be seen in Fig. 4. It was found to decay with the characteristic 85-sec half-life of ^{145}Gd^m. We therefore proposed in Ref. 5 that the γ ray was in fact the previously unobserved groundstate $\frac{3}{2}^+$ to $\frac{1}{2}^+$ transition in ^{145}Gd. This nucleus is the first of the odd-A $N=81$ isotones to have a $\frac{1}{2}$ spin, presumably due to the $s_{1/2}$ neutron orbital. (The spin value was initially deduced[7] from decay data and subsequently determined in a direct measurement.[8]) For the remaining isotones, ^{133}Te–^{143}Sm, the ground state in each instance is represented by the $d_{3/2}$ neutron orbital. The 721.4-keV transition, known to be associated with ^{145}Gd^m, has been shown[9] experimentally to be $M4$ in character. Therefore, as is the case for the other $N=81$ nuclei under discussion, ^{145}Gd^m is the result of a transition between the $h_{11/2}$ and $d_{3/2}$ neutron states. Eppley, McHarris, and Kelly,[9] however, were unable to observe the $d_{3/2}$ to $s_{1/2}$ transition (an upper limit of ≤ 20 keV was set), so that the excitation energies of the $h_{11/2}$ and $d_{3/2}$ states remained unknown. Our suggestion in Ref. 5 was that they were located at 748.7 and 27.3 keV, respectively. This has now been confirmed by Firestone, Warner, McHarris, and Kelly.[10] They not only observed the 27.3-keV γ ray but estab-

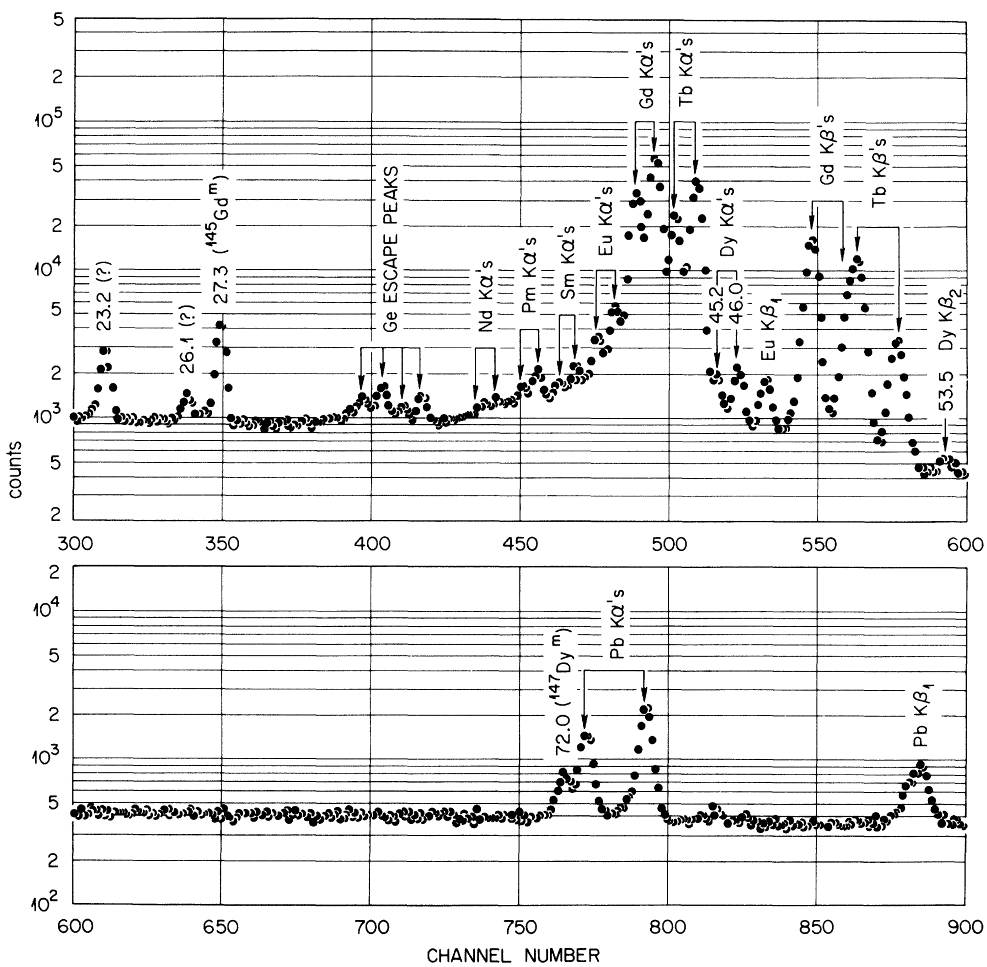

FIG. 4. Spectrum measured with a Ge(Li) x-ray detector. Dysprosium K x rays and the 72.0-keV γ ray, assigned to ^{147}Dy^m, are clearly observed. A 27.3-keV γ ray is also seen. This γ ray was found to decay with the characteristic half-life of ^{145}Gd^m.

lished it to be in coincidence with the 721.4-keV $M4$ transition and determined its multipolarity to be 99.2% $M1$.

The new data concerning the excitation energies of ^{145}Gd levels add substantially to our knowledge of the systematic behavior of the $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$ states in $N=81$ odd-A nuclei. On the basis of these systematics (presented and discussed below) we propose that our ^{147}Dy^m decay data locate the $h_{11/2}$ and $d_{3/2}$ states in ^{147}Dy at 750.7 and 72.0 keV, respectively, above the $s_{1/2}$ ground state. The multipolarities of the 72.0- and 678.7-keV transitions were not determined because of experimental difficulties arising mainly from the low intensities of the two transitions. However, if their multipole orders are assumed to be $M1$ and $M4$, respectively, then their total transition strengths can be calculated from the measured photon intensities. These strengths can then be compared with those of the ^{147}Gd 27.3- and 721.4-keV transitions, calculated once again from photon intensities. As noted in the previous paragraph the multipolarities of the ^{145}Gd transitions have been measured.[9,10] The two sets of intensities were calculated and the following ratios were found: (1) I(27.3 keV)/I(72.0 keV) = 17.7, and (2) I(721.4 keV)/I(678.7 keV) = 18.5. The similarity of the two ratios supports not only the validity of the assumed multipolarities for the ^{147}Dy transitions but also our proposed location of the $h_{11/2}$ and $d_{3/2}$ states in ^{147}Dy.

Excitation energies of the $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$ states for $N=81$ odd-A nuclei are listed in Table I, the data being taken from the present investigation and Refs. 9, 11–18. The location of the three levels in these nuclei is shown graphically in Fig. 5; note that our results for ^{147}Dy fit well into the over-all systematics. (The information in Table I was also presented in tabular form in Ref. 5, but it should be pointed out that some references and footnotes accompanying the table were misplaced.) Included in Table I are the energy differences between the $h_{11/2}$ and $d_{3/2}$ states. It is seen that the new data for ^{145}Gd and ^{147}Dy clearly establish the trend predicted by Silverberg.[19] His shell-model calculations for the $N=81$ isotones showed that the $M4$ transition energies would first increase with increasing atomic number, reach a maximum at about ^{139}Ce, and then begin to decrease.

Attention has been drawn several times in the paper to the low intensity of the ^{147}Dy^m transitions, much lower than had been anticipated at the start of the investigation. This low intensity can be noted from the above comparison with ^{145}Gd^m transitions and from the fact that at 142 MeV the yield of 1.9-min ^{147}Tb was measured to be ~28 times greater than that of ^{147}Dy^m. Two possible explanations can be invoked. The first one has to do with charged-particle emission from the compound system. For nuclei far on the neutron deficient side of β stability, competition of this type is known to depress severely cross sections for reactions where only neutrons are emitted. However, one notes in Fig. 2 that the ^{148}Dy and ^{148}Tb yields are not that different; if half-life differences are accounted for then the ^{148}Tb yield is only ~30% larger than that of ^{148}Dy. A second explanation is that a large fraction of ^{147}Dy^m decays directly to the ^{147}Tb 1.9-min $h_{11/2}$ isomeric state[2] via the β transition $\nu h_{11/2} \rightarrow \pi h_{11/2}$. As the atomic number of the $N=81$ nuclei increases, so does the percentage of direct decay vs the $M4$ transition, from ≤ 0.01 for ^{141}Nd^m to 4.7% for ^{145}Gd^m (see Ref. 9). This is due to the increased occupancy of the $h_{11/2}$ orbital by

TABLE I. Excitation energies of neutron states in $N=81$ nuclei.

Nucleus	Excitation energy (keV) $s_{1/2}$	$d_{3/2}$	$h_{11/2}$	Energy difference (keV) $h_{11/2}$-$d_{3/2}$
^{133}Te	?	0	334.14 ± 0.07 [a]	334.14 ± 0.07 [a]
^{135}Xe	?	0	526.5 ± 0.3 [b]	526.5 ± 0.3 [b]
^{137}Ba	281 ± 5 [c]	0	661.638 ± 0.019 [d]	661.638 ± 0.019 [d]
^{139}Ce	254.7 ± 0.3 [e]	0	754.4 ± 0.7 [f]	754.4 ± 0.7 [f]
^{141}Nd	193.7 ± 0.3 [g]	0	756.5 ± 0.7 [f]	756.5 ± 0.7 [f]
^{143}Sm	108 [h]	0	754.4 ± 0.7 [f]	754.4 ± 0.7 [f]
^{145}Gd	0	27.3 ± 0.1 [i]	748.7 ± 0.5 [i]	721.4 ± 0.4 [j]
^{147}Dy	0	72.0 ± 0.1 [i]	750.7 ± 0.3 [i]	678.7 ± 0.2 [i]

[a] Reference 11.
[b] Reference 12.
[c] Reference 13.
[d] Reference 14.
[e] Reference 15.
[f] Reference 16.
[g] Reference 17.
[h] Reference 18. No error limits given by the authors.
[i] Present investigation.
[j] Reference 9.

proton pairs as the $(g_{7/2}-d_{5/2})$ proton subshell is filled.

The amount of direct decay to ^{147}Tb is of interest since one could then calculate the ^{147}Dy^m $M4$ matrix element for comparison with other isomeric transitions in the series. An experimental determination proved impossible. An initial growth period was not observed in the decay curve of ^{147}Tb γ rays, even when the bombardment time was diminished to 30 sec. The indication is that independent production of ^{147}Tb is substantial enough to obliterate any evidence of a parent-daughter relationship; the precise amount, however, cannot be determined. In the same experiments the decay of the terbium K x rays was followed, but a 59-sec component, due to ^{147}Dy^m electron capture (EC) decay, was not observed. The dominant activity had a half-life slightly greater than 3 min, presumably due to ^{148}Dy. The absence of a 59-sec component cannot be taken to mean that there is no direct decay to ^{147}Tb because the ^{147}Dy Q_{EC} is estimated[20] to be ~7.3 MeV, and positron decay should predominate over electron-capture. (The Q_{EC} for ^{145}Gd^m direct decay to the $h_{11/2}$ state in ^{145}Eu is ~5.7 MeV, and the β^+/EC ratio has been measured[9] to be about 3 : 1.) The annihilation radiation peak observed in our experiments unfortunately encompasses the positron decay of many neighboring nuclei; decay-curve analysis of this peak, without chemical or mass separation, cannot produce meaningful results.

It was therefore decided to see if theoretically predicted cross sections could shed some light. Calculations were made for ^{14}N bombarding energies from 80 to 160 MeV using the statistical-model code ALICE.[21] The code is essentially the compound-nucleus program of Blann (see, e.g., Ref. 22) modified to include fission competition in the deexcitation process. An over-all agreement was found between the predictions and our experiment data. For example, the experimental yield curves shown in Fig. 2 and the predicted excitation functions for the same reactions were found to peak, within a few MeV, at the same incident energies. Another example of the general agreement between the calculations and experiment is as follows. The ratios of maximum cross sections calculated for the production of ^{147}Tb, ^{148}Tb, and ^{148}Dy were 1.15 : 0.78 : 1.00, respectively; the corresponding experimental ratios were 1.3 : 1.3 : 1.0. The point of most interest to the present discussion, however, is that the ALICE calculations, in agreement with the data, predict a very low ^{147}Dy yield. This can be noted from the following list of peak cross sections for ^{141}Pr(^{14}N, xn) products: ^{150}Dy (345 mb), ^{149}Dy (242 mb), ^{148}Dy (146 mb), ^{147}Dy (14.9 mb), and ^{146}Dy (2.3 mb). The sharp decrease in the predicted yield between ^{148}Dy and ^{147}Dy can be understood in terms of increased charged-particle emission. Nevertheless, the calculated (^{147}Tb/^{147}Dy) ratio at the peak of both production cross sections is only 11.3, i.e., much less than the experimental value of ~28 mentioned earlier. This difference suggests that charged-particle competition alone cannot account for the low intensity of the 678.7-keV transition; instead, some significant portion

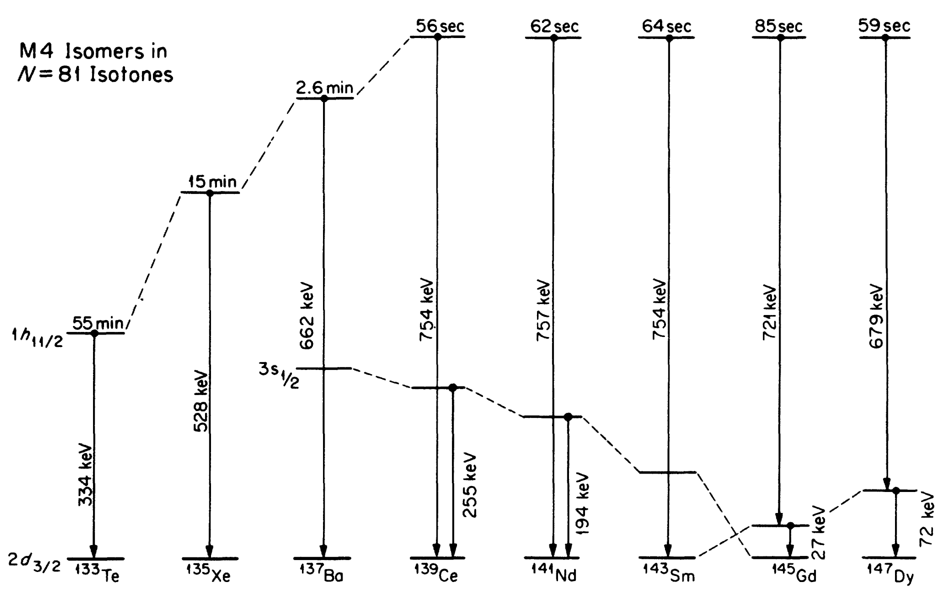

FIG. 5. Location of $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$ neutron states in $N = 81$ odd-A isotones. The new data obtained for ^{145}Gd and ^{147}Dy establish the trend predicted by Silverberg (Ref. 19) for these nuclei; the $M4$ energy should peak at about ^{139}Ce.

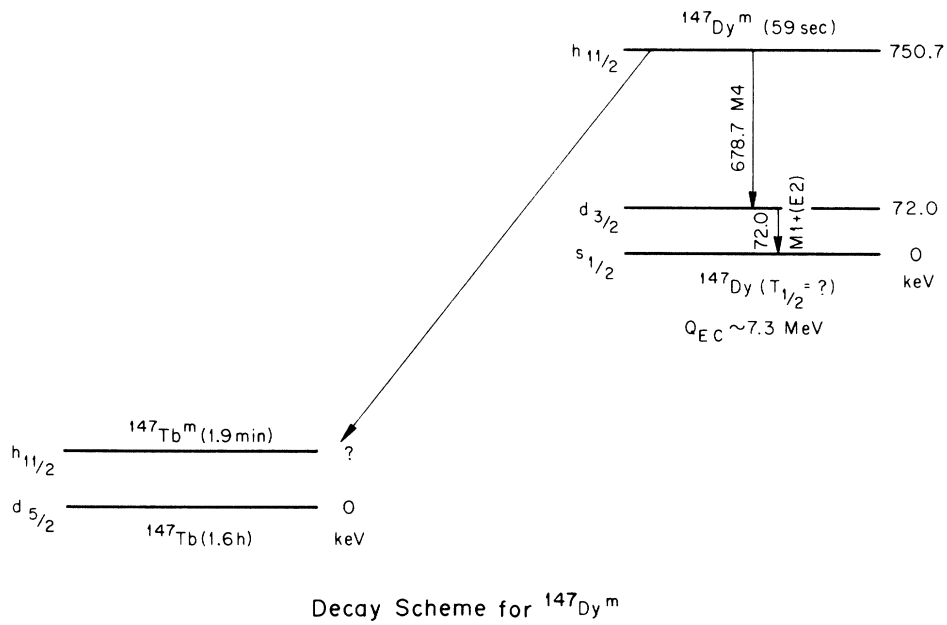

FIG. 6. Proposed ^{147}Dy^m decay scheme. As discussed in the text, statistical model calculations (Ref. 21) indicate that about 60% of ^{147}Dy^m decay proceeds direct to the $h_{11/2}$ state in ^{147}Tb, via the β transition $\nu h_{11/2} \to \pi h_{11/2}$.

of ^{147}Dy^m decay proceeds directly to ^{147}Tb.

The proposed ^{147}Dy^m decay scheme is shown in Fig. 6. If we take the calculated (^{147}Tb/^{147}Dy) ratio at face value then the experimental ratio of ~28 indicates that only about 40% of the ^{147}Dy^m decay strength is represented by the 678.7-keV transition. The partial half-life for the $M4$ isomeric decay would, in that case, be approximately 2.5 min, i.e., similar to the 2.6-min half-life of ^{137}Ba^m, whose $M4$ transition energy is 662 keV (see Fig. 5). The $M4$ radial matrix elements $|M|^2$ for the two isomers would then be quite similar. Firestone *et al.*,[10] who were familiar only with an abstract,[23] determined the ^{147}Dy^m $|M|^2$ value using the total 59-sec half-life. The resultant matrix element is more than twice that of ^{139}Ce^m, ^{141}Nd^m, ^{143}Sm^m, and ^{145}Gd^m and almost twice that of ^{137}Ba^m. They thus concluded[10] that the turnover in the $|M|^2$ vs A curve for the $N=81$ nuclei, as had been predicted in Ref. 9, did indeed occur at $A=147$. Clearly, from the above discussion such a conclusion is premature.

IV. CONCLUSION

To conclude, two observations should be made. First, the likelihood of finding the $M4$ transition in ^{149}Er with experiments similar to these would be expected to be very improbable both because of a low production cross section and an increase in the amount of direct decay to its EC/β^+ daughter, ^{149}Ho. Second, a careful examination of singles and coincidence spectral data did not reveal γ rays that could be ascribed to ^{147}Dy^g decay; instead, all reasonably intense peaks were accounted for by known radioactivities. This result is not surprising. Independent production of the ^{147}Dy ground state in a heavy-ion reaction should be small in comparison with the amount produced via isomeric decay from the $h_{11/2}$ state. The isomeric transition to begin with is low in intensity when compared with the yields of neighboring nuclei. Further, the ^{147}Dy^g decay is very probably fragmented by β transitions proceeding to several final states in ^{147}Tb, as is the case for ^{145}Gd^g decay (see, e.g., Ref. 7). Therefore, the identification of ^{147}Dy^g and the subsequent investigation of its decay scheme will undoubtedly require not only more intense heavy-ion beams but mass separation as well.

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