# Decay Scheme of $Mo^{91}$†

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Gamma-radiation energies were measured at 1540±20, 1210±30, and 658±3 kev, all of which decay with the known 65.5-second half-life. Three positron groups were found to be involved in the same decay. The end-point energies of these were found to be 3.99±0.05, 2.78±0.10, and 2.48±0.10 Mev. No gamma radiation was found decaying with the longer 15-minute half-life. A single positron group with this longer half-life was found to have an end point of 3.44±0.03 Mev. The long-lived positrons represent a transition between the ground states of $Mo^{91}$ and $Nb^{91}$, whereas the 65-second positrons proceed from a 658-kev excited state of $Mo^{91}$ to three excited states in $Nb^{91}$, the lowest of which is the well-known 104-kev excited state. The two higher excited states decay by gamma emission to this state.

## I. INTRODUCTION

MOLYBDENUM-91 has been found by several investigators to exhibit two decay activities, one at about 15.5 minutes[1–9] (15.5±0.2 minutes[8]), and one at about 65 seconds[6–8] (65.5±2 seconds[8]). Duffield and Knight,[7] employing absorption techniques, observed positrons of maximum energy 3.7 Mev associated with the 15-minute decay, and positrons of maximum energy 2.6 Mev associated with the 65-second decay. The same workers reported a 0.3-Mev gamma ray (also on the basis of absorption methods) exhibiting the shorter decay period.

More recent work[9,10] has indicated the presence of three gamma rays accompanying the 65-second decay. Axel *et al.*[9] have measured these energies, by means of scintillation techniques, to be 650±15 kev, 1.22±0.03 Mev, and 1.55±0.03 Mev. No evidence is given by these workers for the existence of a 0.3-Mev gamma ray.

A detailed investigation of the radiations from $Mo^{91}$ decay was undertaken by the present authors in order to attempt to clear up questions regarding the true decay scheme of $Mo^{91}$. The case is of special interest also in view of the position of the isotopes involved, ${}_{42}Mo_{49}{}^{91}$ and ${}_{41}Nb_{50}{}^{91}$, with respect to the magic number 50 for both the neutrons and protons.

## II. APPARATUS

Certain of the activities employed for calibration purposes were obtained from the U. S. Atomic Energy Commission. The remaining activities and sources were produced by means of $(\gamma,n)$ reactions with the University of Illinois 22-Mev betatron employing the probe technique.[11]

The bulk of the measurements associated with the 65-second activity were made by means of a ten-channel scintillation spectrometer.[12] In some instances this was supplemented by a single-channel scintillation device of adjustable width and sensitivity. This latter procedure has been discussed previously.[13]

The particles associated with the 15.5-minute activity were measured with a double-focusing magnetic spectrometer of the Siegbahn type, having a mean radius of 15 cm. The detector was a double-beaded, end-window Geiger counter, the window consisting of Zapon film of thickness somewhat less than 100 $\mu g/cm^2$. For the energies involved in these measurements no corrections for the window thickness were necessary. The counter was filled with argon at 2 cm pressure and alcohol at about 1 cm, the latter being kept constant by maintaining the counter system open to a vessel of alcohol immersed in a melting ice bath. The presence of the bead at the middle of the counter wire makes possible the discrimination against events occurring in the wall of the counter and favors detection of particles entering through the window parallel to the axis of the counter. For the latter cases the particles are able to trip both regions of the counter, thus giving a pulse twice as large as those produced by events which discharge but one region. Consequently, background can be reduced in this manner. These techniques have been consistently employed both in this laboratory in the past and also by others.

## III. DATA

Molybdenum activities were prepared by irradiating the unseparated metallic sheet. For particle and x-ray work material of surface density 17 $mg/cm^2$ was used, and for work involving only gamma rays the surface density was 50 $mg/cm^2$.

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### 1. 15-Minute Positrons

Figure 1 shows a Kurie plot of the 15-minute positron group, the data having been obtained with the 255° magnetic spectrometer. On the basis of an energy calibration with the positrons of $Cu^{62}$, taken as having an end-point energy of 2.91 Mev, the maximum energy of the $Mo^{91}$ particles was found to be $3.44\pm0.03$ Mev. From the data it would appear that the spectrum is simple. This is consistent with the fact that no gamma radiation having the same half-life was found.

### 2. Photon Spectra

The gamma radiation was observed with the ten-channel scintillation spectrometer, employing a NaI(Tl) crystal ($1\frac{1}{2}$-in. diam$\times$1 in.). The top curve in Fig. 2 represents a composite spectrum of the two regions, 350 to 950 kev, and 800 to 1650 kev. The two sets of data were normalized in the 700–1000 kev region. The data were calibrated by means of the 661-kev gamma ray following the $Cs^{137}$ decay (not shown in figure) and by the 1.28-Mev radiation following the decay of $Na^{22}$. For those points in which the statistical errors are not indicated the latter are equal to or less than the breadth of the point symbol. Three gamma rays having a decay half-life of approximately a minute were found. None were found which could be attributed to the 15-minute activity.

The energy of the gamma ray which we shall designate as $\gamma_1$ was found to be $658\pm3$ kev. For the precise energy determination of $\gamma_1$ it was carefully compared at high dispersion (data not shown) with the 661-kev Cs radiation and the 650-kev gamma ray associated with the decay of $Os^{185}$. It was evident that the $\gamma_1$ peak lay between these two energies, and somewhat closer to the Cs peak than to the Os peak. The value of 658 kev just quoted represents the mean of the results of six determinations.

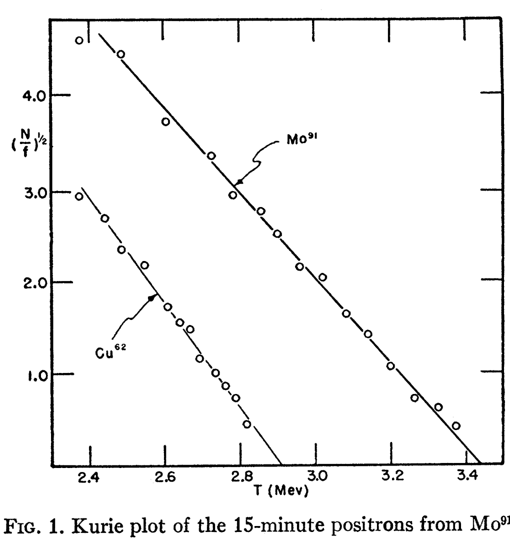


FIG. 1. Kurie plot of the 15-minute positrons from $Mo^{91}$.

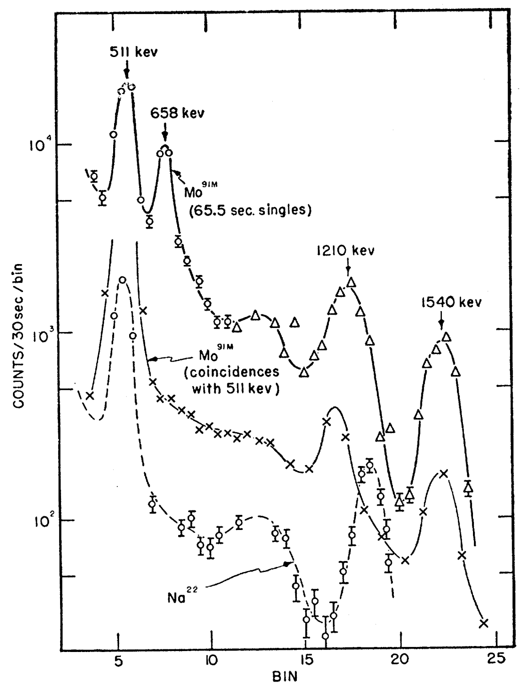


FIG. 2. Pulse-height spectrum of gamma rays from $Mo^{91m}$ with a NaI(Tl) crystal. The upper curve is the spectrum of single counts; the middle curve is the gamma spectrum which is in coincidence with annihilation radiation.

The energies of the remaining two gamma rays, $\gamma_2$ and $\gamma_3$, were found to be $1540\pm20$ kev for $\gamma_2$ and $1210\pm30$ kev for $\gamma_3$.

The relative intensities of the three gamma rays and annihilation radiation were measured. The efficiency curve of the system, geometry and NaI(Tl) crystal, was empirically determined by observing, for other activities, pairs of radiation peaks the relative intensities of which are known. In the case of $Na^{22}$ the true relative intensity of $\gamma_{1280}:\gamma_{511}$ was taken to be 1.2:2.0, assuming 10% orbital capture. This follows closely the result of Kreger.[14] $Os^{85}$ and $Co^{60}$ were also employed, the ratio of 650-kev to 880-kev radiation in $Os^{85}$ being taken to be 85:15,[15] and the 1.17-Mev and 1.33-Mev radiation of $Co^{60}$ being assumed of equal intensity.[16] The efficiency curve was extrapolated to higher energy by comparison with a similar curve found by Kahn and Lyon[17] for a crystal of this size.

For the 65-second $Mo^{91}$ activity the relative intensity of $\gamma_3:\gamma_2=0.68\pm0.1$. The apparent intensity of the annihilation radiation to the 658-kev radiation, corrected for crystal efficiency, was found to be $\gamma_{511}:\gamma_1=1.4\pm0.2$. Thus the intensity ratio of positrons to the 658-kev radiation was $\beta^+:\gamma_1=0.7\pm0.1$.

[14] W. E. Kreger, Phys. Rev. **96**, 1554 (1954).
[15] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[16] K. Way, *Nuclear Data*, National Bureau of Standards Circular 499 (U. S. Government Printing Office, Washington, D. C., 1950).
[17] B. Kahn and W. S. Lyon, Nucleonics **11–11**, 61 (1953).

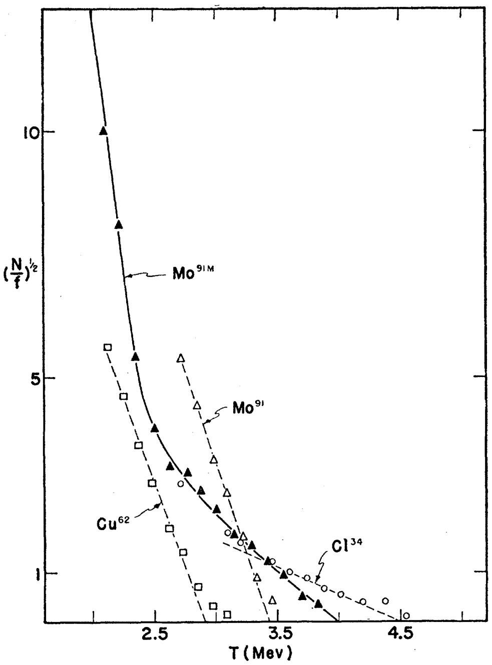


FIG. 3. Kurie plot of the 65-second positrons from $Mo^{91m}$.

A large number of x-rays were found. These were attributed both to orbital capture transitions and to conversion in $Mo^{91}$. The ratio of the intensity of $K$ x-rays to 658-kev gammas was determined by comparison with $Cs^{137}$, employing fixed geometry and a thin crystal for x-ray detection. $Zr^{89}$ and $Ag^{106}$ were also checked in this manner, the results obtained being in fair agreement with those of Shore[12] and Bendel.[18] In the present experiments, employing an x to 661-kev gamma ratio for $Cs^{137}$ of 0.10, the result was $x:\gamma_1=0.15\pm0.05$.

### 3. 65-Second Positrons

The 65-second positrons were observed with the ten-channel scintillation spectrometer, employing both a plastic Sintilon crystal with a small hole drilled in it and an anthracene crystal. The results were comparable in the two cases and are depicted in the form of a Kurie plot in Fig. 3. The data were calibrated, as shown, by means of the end points of $Cl^{34}$, $Cu^{62}$, $Zn^{63}$, and 15-minute $Mo^{91}$. The data indicated that the positron spectrum is complex, with a maximum end point at $3.99\pm0.05$ Mev. A detailed analysis of the data revealed the higher energy group to have about 15% of the intensity of the lower energy particles.

### 4. Coincidence Experiments

Two lower energy positron groups, also decaying with the 65-second half-life, were found by measuring the particles in coincidence with gamma radiation of specified energy. In this experiment the single-channel analyzer [plus a NaI(Tl) crystal] was used for the detection of the gammas. The ten-channel instrument, with a plastic scintillator, was employed for the particles. Figure 4 shows Kurie plots of the particles, the energy calibration being obtained with the positrons from $Cu^{62}$ and from 15-minute $Mo^{91}$. The results indicate that $\gamma_2$ is in coincidence with particles of end point $2.48\pm0.10$ Mev, and $\gamma_3$ is in coincidence with a group having an end-point energy of $2.78\pm0.10$ Mev.

A second coincidence type measurement involved examining the energies of the gamma rays in coincidence with 511-kev radiation. NaI(Tl) crystals were employed for this, in conjunction with the two pulse-height analyzers. The results are shown in Fig. 2, and indicate that both $\gamma_2$ and $\gamma_3$ follow positron decays. A careful examination of data taken in the region of 600 to 800 kev failed to disclose any 658-kev gamma rays to be in coincidence with annihilation radiation.

Gamma-gamma coincidences were looked for in a possible cascade of $\gamma_2$ and $\gamma_3$. A few prompt coincidences were found involving radiation of energy in the range of 700 kev to 2000 kev. However, a careful comparison with the case of $Co^{60}$ showed that no more than about 5% of $\gamma_2$ and $\gamma_3$ could be involved in a possible cascade. In view of the presence of high-energy positrons, these coincidences were attributed, instead, to bremsstrahlung.

### 5. Conversion Electrons

Conversion electrons were found which possessed a half-life of approximately a minute. A single peak at about 640 kev was found. This was attributed to the 658-kev transition. None were detected which could be associated with the 1540- and 1210-kev transitions.

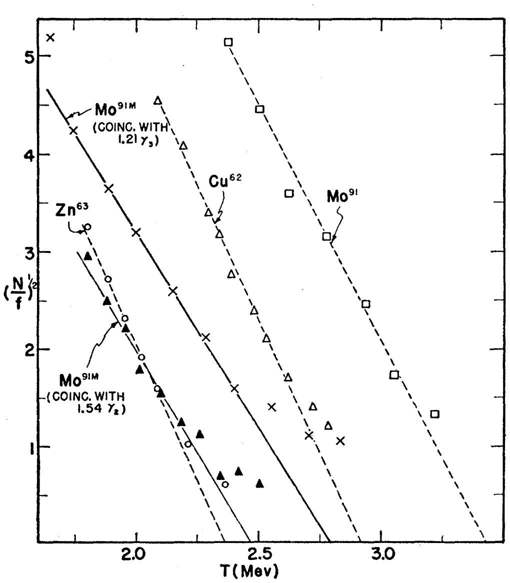


FIG. 4. Kurie plot of the 65-sec positrons in coincidence with gamma rays of specified energy.

[18] Bendel, Shore, Brown, and Becker, Phys. Rev. 90, 888 (1953).

The total conversion coefficient was determined with the use of two scintillation detectors, the ten-channel analyzer in conjunction with a thin stilbene cyrstal collecting the particles, and the single-channel analyzer the gamma rays. Calibration of the geometry was done with Cs$^{137}$. When the value for Cs was taken to be 0.11, the result for the 658-kev radiation of Mo$^{91}$ was $0.055 \pm 0.015$.

## IV. DISCUSSION

Figure 5 shows a tentative level scheme embodying the results of the present investigation. The relative intensities were estimated from the measured values $\beta^+/\gamma_1 = 0.7$, $e_1/\gamma_1 = 0.055$, $\gamma_3/\gamma_2 = 0.68$, and $\beta_4{}^+ = 0.15 \times (\beta_2{}^+ + \beta_3{}^+)$, and from the theoretical estimates of the relative intensity of orbital capture to positron decay, assuming allowed transitions and following Feenberg and Trigg.[19] In the cases of the positrons, the upper number is the kinetic energy in Mev, while the numbers in parenthesis are the percentages of the decays of parent level and the $\log ft$ value.

Considerations of the energies found for the various radiations favor very strongly the order of the levels as presented in the figure. The partial lifetime of $\gamma_1 + e_1$, found from the relative intensities listed in Fig. 5, together with the total conversion coefficient measured

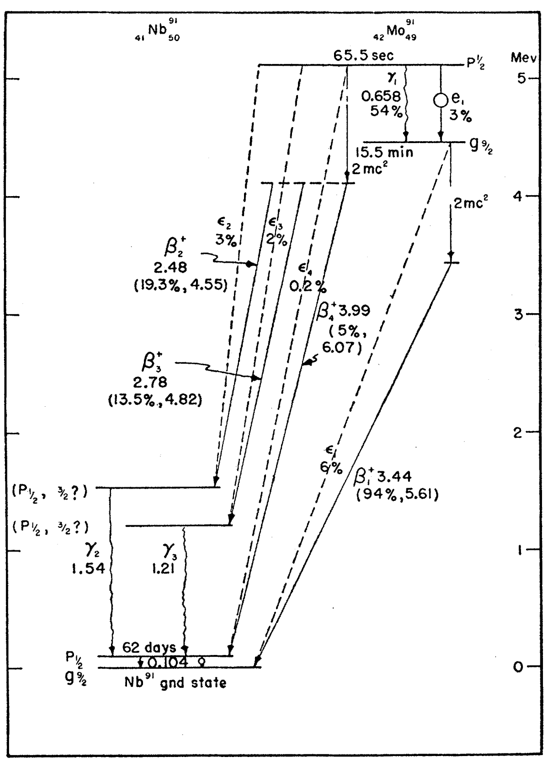


FIG. 5. Level scheme involved in the decay of Mo$^{91}$ and Mo$^{91m}$ to Nb$^{91}$ and Nb$^{91m}$.

[19] E. Feenberg and G. L. Trigg, Revs. Modern Phys. **22**, 399 (1950).

in the present investigation, are consistent with designating the transition to be $M4$. That the 62-day, 104-kev gamma ray is also $M4$ is due to other sources.[20] The spin-orbit coupling shell model suggests that in this region of the periodic table an isomeric pair of levels giving rise to an $M4$ transition shall have the single-particle configurations $g_{9/2}$ and $p_{1/2}$. If we follow this suggestion for the pair of levels in Mo$^{91}$, the work of Axel, Fox, Parker,[9] and of Brolley[21] indicates the $g_{9/2}$ level to be the lower of the two.

Hayward *et al.*,[22,23] at the 1954 Chicago meeting of the American Physical Society, reported that the 122-Mev gamma ray in Zr$^{91}$, previously known[24] to follow the decay of Y$^{91}$, also follows the decay of the 104-kev level of Nb$^{91}$. The measured spin of the ground state of Zr$^{91}$ is 5/2, in accord with a $d_{5/2}$ shell-model level. Also, since the Y$^{91}$ ground state has been designated[20,23] $1/2^-$, and the 1.22-Mev state of Zr$^{91}$ as either $1/2^-$ or $3/2^-$, owing to the observed 0.33-Mev[23,24] beta transition from Y$^{91}$, the work of Hayward *et al.* favors the assignment of $1/2^-$ to the 104-kev state of Nb$^{91}$.

The details and energetics of the Mo$^{91}$—Nb$^{91}$ decay scheme ascertained in the present work are in good agreement with both the conclusions of Axel and Brolley, mentioned above, for Mo$^{91}$, and those of Hayward *et al.* and of Bunker *et al.*[24] for Zr$^{91}$. In view of these assignments in Mo$^{91}$ and Zr$^{91}$, we designate the ground state of Nb$^{91}$ to be $g_{9/2}$ and the 104-kev state to be $p_{1/2}$.

It is to be noticed that the high-energy positron $\beta_4{}^+$, involved in the transition between the two $p_{1/2}$ levels, has a rather high $ft$ value. This behavior has been observed before for similar transitions, also designated as allowed, in Y$^{87}$ to Sr$^{87}$ (studied by Mann and Axel[25]), Kr$^{85}$ to Rb$^{85}$ (studied by Sunyar *et al.*[26]), and Zr$^{89}$ to Y$^{89}$ (studied by Shore *et al.*[12]). All of these examples, including the present one, are in the region of the periodic table such that the number of protons and the number of neutrons in each isotope involved is in the range 36 to 50. The notion of configurational mixing has been employed in order to account for these.

The transitions $\beta_2{}^+$ and $\beta_3{}^+$, shown in Fig. 5, are considered to be allowed because of the low $ft$ values. Accordingly, for these transitions to be consistent with Gamow-Teller selection rules, the two high levels in Nb$^{91}$ must be designated as $p$ states. In Nb$^{91}$, according to the shell model, the three protons beyond the closed subshell at 38 nucleons would chiefly determine the

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[21] J. E. Brolley, Jr., Phys. Rev. **89**, 877 (1953).
[22] Hayward, Hoppes, and Ernst, Phys. Rev. **98**, 231(A) (1955).
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[26] Sunyar, Mihelich, Scharff-Goldhaber, Goldhaber, Wall, and Deutsch, Phys. Rev. **86**, 1023 (1952).

character of low-lying levels. Accordingly, the two lower levels are thought to be described by the proton configurations $38(p_{1/2})^2(g_{9/2})$ or $38(g_{9/2})^3$, for the ground state, and $38(p_{1/2})(g_{9/2})^2$ for the first excited state. For the two upper states it is possible to achieve a $p_{3/2}$ designation for both by assuming two distinct linear combinations of the single-particle configurations $34(p_{3/2})^3(p_{1/2})^2(g_{9/2})^2$ and $34(p_{3/2})^3(g_{9/2})^4$.

Figure 6 shows a plot, following the method of Goldhaber and Hill,[20] of the odd-$A$ niobium isotopes. The diagram shows energy differences between the $p_{1/2}$ and the $g_{9/2}$ states as a function of neutron number. The curves correspond to energies of the $p_{1/2}$ states whereas the $g_{9/2}$ levels are on a single horizontal line. Thus for a group of isotopes like those of Nb, in which the $p_{1/2}$ curve is above the horizontal line, the $g_{9/2}$ level is the ground state. It is interesting to notice that the minimum is now to be found at a neutron number of 52 rather than the previously assumed[20] point at 50 neutrons. It is interesting to speculate on the possible existence of an isomeric state in $_{41}Nb_{48}{}^{89}$. If the trend continues as shown by the dotted portion of the curve one might expect such a level at about 300 or 400 kev, with a half-life of several hours, depending on the competing modes of decay. Some indication of such a state has been found by Mathur *et al.*[27] At the other end of the curve, the designations shown for $Nb^{95}$ and $Nb^{97}$ can now be regarded to rest on a firmer basis because of the recent measurement of the spins of $Mo^{95}$ and $Mo^{97}$ by Owen and Ward.[28]

It is of interest to investigate the cases of $Y^{85}$ and $Y^{93}$ for a possible crossover of the $p_{1/2}$ and $g_{9/2}$ levels. Evidence has been cited for a 0.7-Mev transition in the latter isotope.[29] The curve of technetium is of interest especially at the lower end. $_{43}Tc_{50}{}^{93}$ (not shown) has been reported to have a 390-kev transition, identified

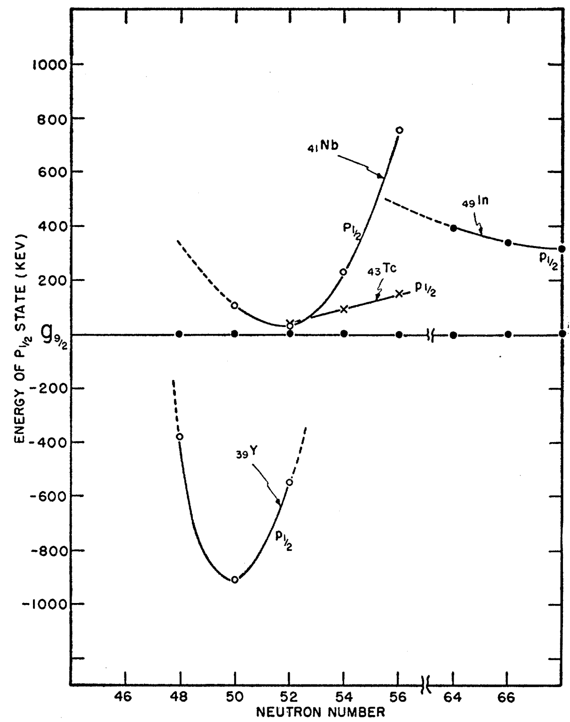


FIG. 6. Energy difference between $2p_{1/2}$ levels and $1g_{9/2}$ levels for odd-proton isotopes in the range $Z=39$ to 49, plotted as a function of neutron number.

as $M4$ by measurement of conversion electrons.[30] The exact level assignment is uncertain. However, further work should lend more certainty since this nucleus decays to $Mo^{93}$ which subsequently decays to $Nb^{93}$, the ground state of which has been measured to be 9/2. Whichever state in $Tc^{93}$ is lower, the 390-kev difference is a deviation from the trend of the Tc curve.

In the case of indium, extrapolation of the curve suggests an isomeric state in $In^{111}$ at approximately 450 to 500 kev with a half-life of the order of hours.

A final feature to be noted in the figure is that the only curve with a minimum at 50 neutrons is that of yttrium.

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[27] Mathur, Hyde, Levine, and Kofstad, Phys. Rev. **97**, 117 (1955).
[28] J. Owen and I. M. Ward, Phys. Rev. **102**, 591 (1956).
[29] N. E. Ballou, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 695.
[30] Bernas, Beydon, Papineau, Compt. rend. **238**, 791 (1954).