# Excitation Functions and Yield Ratios for the Isomeric Pairs $Br^{80,80m}$, $Co^{58,58m}$, and $Sc^{44,44m}$ Formed in $(p,pn)$ Reactions*

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(Received August 30, 1955; revised manuscript received December 19, 1955)

Excitation functions and yield ratios for isomeric pairs have been measured from near threshold to 100 Mev for three $(p,pn)$ reactions. The results are interpreted in terms of the spins of the states involved, the angular momentum of the bombarding particle, competition between compound nucleus formation and the knock-on mechanism and the level density in excited nuclei as a function of spin.

## INTRODUCTION

NUCLEAR reactions are usually considered as proceeding by compound nucleus formation[1] at energies <30 Mev and by the knock-on mechanism[2] at energies >100 Mev. The two mechanisms can lead to the same intermediate nuclei with about the same excitation energy but with different distributions in the values of angular momentum. This should result in a different distribution in the yields of end products when they differ greatly in spin, as do nuclear isomers. A study of the excitation functions and relative yields of isomeric states produced by a simple reaction from threshold to 100 Mev might help show up changes in the reaction mechanism with energy.

Since most of the data in the literature[3–7] on isomer production have been obtained in low-energy reactions, no coherent picture of the variation of relative yields with energy or of the effect of reaction mechanism exists. At low energies the spins of the initial nucleus and of the isomers are a major factor in determining the ratio of the yields.[3,4] That isomer will be favored whose spin is nearest that of the target nucleus. At higher energies where the compound nucleus is formed in a wider range of spin states this effect might be expected to disappear, and it has been suggested that the yield ratio should then approach the ratio of the statistical weights of the isomeric states.[4] However, it has been shown that, in at least one case, the statistical weight ratio presents no limiting value.[8]

## EXPERIMENTAL

### Bombardment Procedure

The excitation functions were obtained using the stacked foil technique in which several targets are interspaced with absorbers to allow for simultaneous bombardment over a wide energy interval. The majority of the bombardments were made in the internal scattered beam of the Harvard cyclotron with 180° focusing in the cyclotron magnetic field. The details of this method have been described by Hintz and Ramsey.[9] Initial proton energies of 73 and 100 Mev were used. The value of this energy was checked before each bombardment by measuring the $C^{12}(p,pn)C^{11}$ excitation function with the same target geometry and comparing it with the known function for this reaction.[9–11] The energy of the protons at each target in the stack was calculated from the range-energy curves of Aron, Hoffman, and Williams.[12]

The spread in energy of the proton beam at the face of the target stack is 0.6 or 0.9 Mev depending upon whether $\frac{5}{16}$- or $\frac{7}{16}$-inch diameter targets were used. This inhomogeneity in the energy of the proton beam increases with depth in the target stack becoming about 6 Mev at a nominal proton energy of 14 Mev and thus obscuring the excitation function near the threshold. For this reason two cobalt bombardments were made on the Berkeley linear accelerator[13] whose incident proton energy was known to be 31.5±0.2 Mev.

Absolute cross sections were determined for the reactions by including aluminum foils in the target

* Supported by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

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[10] Aamodt, Peterson, and Phillips, Phys. Rev. **88**, 739 (1952).

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[12] Aron, Hoffman, and Williams, University of California Radiation Laboratory Report UCRL-121, 1949 (unpublished).

[13] The authors are grateful for the cooperation and assistance of Dr. E. K. Hyde of the University of California Radiation Laboratory in arranging the linear accelerator bombardments.

stacks and monitoring the beam by means of the $Al^{27}(p,3pn)Na^{24}$ reaction as determined by Hintz and Ramsey,[9] but corrected to the more recent results for the $C^{12}(p,pn)C^{11}$ reaction.[11]

The bromine targets consisted of sodium bromide pressed into wafers $\frac{5}{16}$ inch in diameter having a surface density of about 70 mg/cm². The scandium targets were of scandium oxide[14] which had been carefully purified from residual calcium, thorium, zirconium and rare earths, and was pressed into $\frac{7}{16}$- or $\frac{5}{16}$-inch diameter wafers which had a surface density of 20–40 mg/cm². Each wafer was packed into a flat aluminum capsule for bombardment. The cobalt targets for the cyclotron bombardments were metal foils $\frac{7}{16}$ inch in diameter with a surface density of 50–60 mg/cm². These were placed in slightly recessed copper foil holders for bombardment. The linear accelerator targets consisted entirely of stacks of very thin (~10 mg/cm²) cobalt metal foils held in a recessed brass block and covered with a thin cobalt window.

## Chemical Procedures

The bromine was precipitated as silver bromide in the presence of hold-back carriers of near-by elements and deposited on a filter paper disk for counting.

The cobalt samples were obtained from the cobalt targets by a variety of solvent extraction procedures. Final purification was made by precipitation with $KNO_2$. For the cyclotron targets, counting samples were made by electroplating the cobalt onto copper plates. The linear accelerator targets were redissolved and placed in vials for counting in a well-type scintillation counter and the yields were determined colormetrically.

The scandium was precipitated twice as the hydroxide, redissolved, and placed in vials for counting in a scintillation counter. Chemical yields were determined by evaporating the solutions to dryness and igniting to the oxide.

## Counting Procedures

Figure 1 gives the decay schemes[15–17] of the three isomeric pairs studied. In determining the relative yields of genetically related isomers, it is advantageous to count only the activity of the lower state. Then the relative yields can be determined from the resolution of the decay curves and no counting corrections need be applied. It was possible to use this procedure for bromine and scandium at all energies and for cobalt at energies less than 43 Mev, the threshold for interfering activities.

The 4.5-hour upper state of $Br^{80}$ decays completely

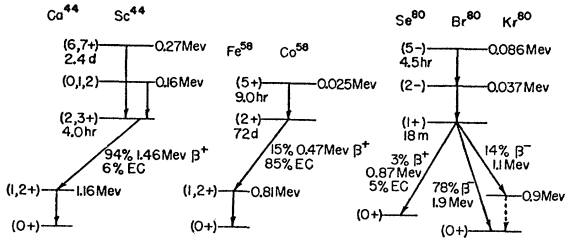


FIG. 1. Decay schemes of $Br^{80}$, $Co^{58}$, and $Sc^{44}$.

to the 18-minute ground state. The $\beta^-$ radiation of the latter was separated from the other bromine activities by means of a simple magnetic bender and counted with a Geiger tube. Although this method worked well for determining the ratio of isomer yields, it was not suitable for determining the excitation function as the counting rate was too sensitive to the distribution of the source. Instead the samples were counted directly with an end-window Geiger tube, and the yield of the 4.5-hour isomer was determined by resolution of the decay curve. The excitation function of the 18-minute isomer was then obtained from the previous ratio.

The cobalt targets from the cyclotron bombardments were counted with an end-window Geiger tube. It was possible to follow the decay of the 9-hour $Co^{58m}$ directly as the cobalt x-rays resulting from the highly converted 25-kev gamma transition had a fairly high counting efficiency. Interference from the 18-hour $Co^{55}$ occurred above 43 Mev, so samples produced above that energy were counted through a 980-mg/cm² beryllium absorber which suppressed the 18-hour activity by a factor of 30 while reducing the $Co^{58m}$ by only 7.3. The total yield of $Co^{58g}+Co^{58m}$ was determined by counting the samples after all the 9-hour activity had decayed to the 72-day ground state. Aluminum absorption curves served to separate the $Co^{58}$ from the 270-day $Co^{57}$ and the 72-day $Co^{56}$.

The targets irradiated in the Berkeley linear accelerator were counted in a well-type NaI(Tl) scintillation counter with the amplifier discriminator so biased as not to count any of the low-energy $Co^{57}$ and $Co^{58m}$ radiations. Thus only the $Co^{58}$ ground-state activity could register. Since the maximum proton energy was 31.5 Mev, there was no interference from $Co^{55}$ and $Co^{56}$.

The $Sc^{44}$ was counted with a NaI(Tl) scintillation counter with the amplifier discriminator so biased as to cut out gamma radiation below 0.9 Mev. This permitted only the 1.16-Mev gamma of the ground state to be counted and eliminated any interference from the 3.0-hour $Ti^{45}$ or the 4.0-hour $Sc^{43}$. Several of the $Sc^{44}$ targets were precipitated, filtered, and counted by an end-window Geiger counter to determine their yield relative to the aluminum monitor targets.

The aluminum monitor foils were counted by Geiger tubes in the same geometry and mounting as their respective targets. All the usual counting corrections

[14] The $Sc_2O_3$ was kindly furnished by Professor G. Wilkinson, Chemistry Department, Harvard University.
[15] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[16] J. W. Blue and E. Bleuler, Phys. Rev. **99**, 659(A) (1955).
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TABLE I. Experimental errors in the cyclotron data. $\Delta\sigma$ (absolute) does not include the possible error in the cross section of the monitor reaction. $\Delta(\sigma_m/\sigma_g)$ refers to counting statistics only.

| | $Br^{80}$ | $Co^{58}$ | $Sc^{44}$ |
|---|---|---|---|
| $\Delta\sigma$ (absolute) | ±18% | ±30% | ±15% |
| $\Delta\sigma$ (relative) | ± 8 | ±10 | ± 4 |
| $\Delta(\sigma_m/\sigma_g)$ | ± 3 | ±10 | ± 3 |

were made with the exception of the one for backscattering. No correction was made for backscattering of $\beta$ radiation since the samples and monitors were always mounted on the same backing material and saturation backscattering is largely independent of energy.[18]

## DISCUSSION

Table I lists the estimated experimental errors for the cyclotron data. The unusually large value of $\Delta\sigma$ (absolute) for $Co^{58}$ is chiefly due to the uncertainty in the counting efficiency. The scatter observed in the linear accelerator data (Fig. 6) is for the most part due to the difficulty in resolving the smaller amount of 9-hour activity remaining after the additional 18-hour period required to transport the target from Berkeley to Cambridge.

There are two additional sources of errors in the cyclotron data at energies <20 Mev. First, the energy spread has increased to several Mev. Since the excitation functions are increasing rapidly with energy in this region, the high-energy part of this spread will have the greatest weight. Second, near the threshold $(n,2n)$ reactions from secondary neutrons become comparable in magnitude to the $(p,pn)$ reactions. The yield from secondary neutrons is from 1–2 percent of the maximum yield of the $(p,pn)$ reactions. In order to minimize this last effect, no points below 15 Mev have been included in the yield ratios.

Inspection of the excitation functions (Figs. 2–4), shows that they have the general shape already observed for such reactions, namely an initial steep rise from threshold to a maximum at 20–30 Mev followed by a more or less sharp fall to a value which then decreases only slowly with increasing energy.

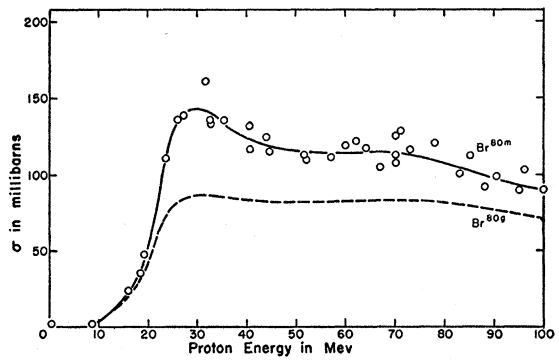


FIG. 2. Excitation functions for the reactions $Br^{81}(p,pn)Br^{80m}$ and $Br^{81}(p,pn)Br^{80g}$.

The plots of the ratio of the cross section for isomer formation to that for ground-state formation $(\sigma_m/\sigma_g)$ *vs* energy are shown in Figs. 5–7. In no case does this ratio approach as a limit that of the statistical weights,[4] $(2I_m+1)/(2I_g+1+2I_i+1)$, where $I_m$, $I_g$, and $I_i$ are the spins of the metastable, ground, and intermediate states, respectively. Neither do they approach values greatly favoring the high-spin state as suggested by an argument in reference 8.

Perhaps the most important factor controlling the variation of the ratio $\sigma_m/\sigma_g$ with energy is the distribution of the angular momentum values of the excited nuclei. At energies <30 Mev, the reaction may be considered as proceeding primarily by the compound nucleus mechanism. If it is assumed that every particle that penetrates the potential barrier will be captured, the distribution of the angular momentum, $J$, of the compound nuclei will be given by

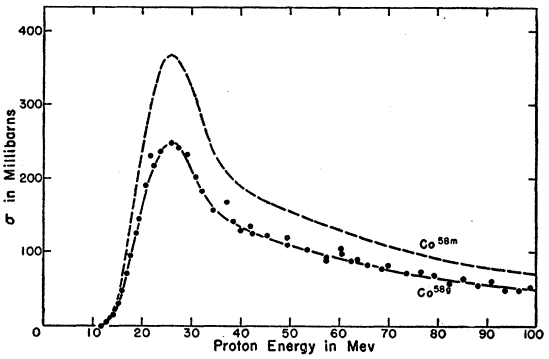


FIG. 3. Excitation functions for the reactions $Co^{59}(p,pn)Co^{58m}$ and $Co^{59}(p,pn)Co^{58g}$.

$$\sigma(J,E)=\pi\lambda^2\sum_{S=I-s}^{I+s}\sum_{l=J-S}^{J+S}g(S)\frac{2J+1}{2S+1}T_l(E),$$

where $T_l(E)$ is the barrier transmission coefficient of a particle with orbital angular momentum $l$ and energy $E$, $S$ is the channel spin, $I$ is the spin of the target nucleus, $s$ is the spin of the particle, and $g(S)$ is the statistical weight of $S$. Figure 8 shows the distribution of $J$ for protons on $Br^{81}$ $(I=3/2)$ and $Sc^{45}$ $(I=7/2)$ and demonstrates the effect of the spin of the target nucleus. The most probable value of $J$ will be near $I$ until $T_l(E)$ for $l\geq I$ becomes large. For a low-spin target nucleus the relative number of states with large values of $J$ increases rapidly with bombarding energy, while for a high-spin target nucleus the rate of increase is much smaller at first.

This initial distribution of angular momentum values will be modified by the de-excitation process which, for the reactions studied, consists of the emission of a

[18] B. P. Burtt, Nucleonics 5, No. 8, 28 (1949).

neutron and a proton followed by a gamma cascade. The spin of the residual nucleus, $I$, is the vector sum of the spin of the compound nucleus, the orbital angular momentum of the particle, and the particle spin. Since a few Mev above the reaction threshold the nucleons may be emitted with fairly high values of angular momentum and the final cascade may consist of several gammas, large changes in the nuclear spin are possible. The decomposition of $J$ may be treated in much the same way as its formation, except that now the effect of the level density of the residual nucleus as a function of spin as well as energy must now be considered. This is very important since otherwise the factor $2I+1$ introduced by the decomposition of $J$ greatly favors the higher spin states. The dependence of the energy level density on angular momentum has been derived on the basis of the Fermi gas model of the nucleus.[19,20] This is

$$\omega_I(E)=\omega_0(E)\frac{I+\frac{1}{2}}{c\tau}\exp[-(I+\tfrac{1}{2})^2/2c\tau],$$

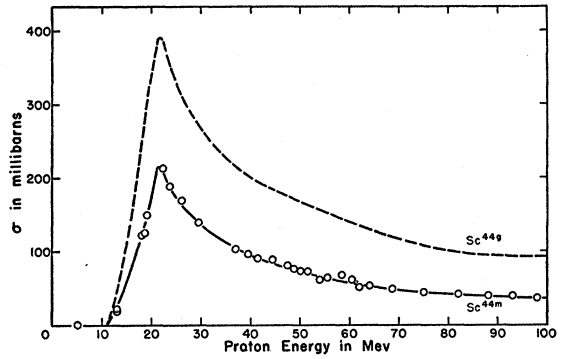


FIG. 4. Excitation functions for the reactions $Sc^{45}(p,pn)Sc^{44m}$ and $Sc^{45}(p,pn)Sc^{44g}$.

where $\tau$ is the nuclear temperature and $c$ is a constant whose value increases with increasing mass number and $\omega_0(E)$ is the total level density. If the nuclear temperature is assumed to be given by $(E/a)^{\frac{1}{2}}$ with the values of $a$ being those given by Blatt and Weisskopf,[1] it can be seen that the most probable value of $I$ increases with increasing mass number and excitation energy. For example, for a mass number of 44 and an excitation energy of 30 Mev the most probable value of $I$ is $\sim 7$ while at 5 Mev it is 3–4. For a mass number of 80 the corresponding values are $\sim 9$ and $\sim 6$. There is experimental evidence which indicates that actual nuclear temperatures may be much lower than the ones used here.[21] This would result in much lower values for the most probable $I$.

The above considerations indicate that the ratio of isomer yields, and particularly the rate of change of this ratio with bombarding energy, is determined largely

[19] H. A. Bethe, Revs. Modern Phys. **9**, 84 (1937).
[20] C. Bloch, Phys. Rev. **93**, 1094 (1954).
[21] P. C. Gugelot, Phys. Rev. **93**, 425 (1954).

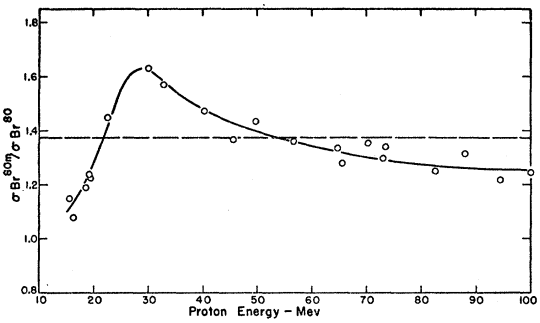


FIG. 5. $\sigma_m/\sigma_g$ for $Br^{80}$. The dashed line represents the ratio of the statistical weights.

by the distribution of angular momentum in the compound nuclei. For the reactions studied here, the deexcitation process will cause the lower spin isomer to be favored to a greater extent than would be indicated by the initial distribution of spins alone, provided that the energy available is several Mev greater than the threshold of the reaction. Thus, in the region beginning a few Mev above the reaction threshold, the compound nucleus mechanism predicts a rapid increase in the relative yield of the high-spin isomer from a $(p,pn)$ reaction with a low-spin target nucleus because of the very rapid increase of the relative number of compound nuclei with high values of $J$ (Fig. 8). For a high-spin target nucleus, the much slower increase in the relative number of compound nuclei with high values of $J$ should result in an even slower increase in the relative yield of the high-spin isomer. This corresponds to the observed results for $Br^{81}$ $(I=3/2)$ and $Sc^{45}$ $(I=7/2)$ as shown in Figs. 5 and 7. $Co^{59}$ should behave in much the same way as $Sc^{45}$ since the two nuclei have the same spin. The value of $\sigma_m/\sigma_g$ appears to be constant above 16 Mev (Fig. 6), although the scatter in the linear accelerator data is sufficient to hide any small variations. The cyclotron data does appear to indicate an initial increase but only one point is outside experimental error.

The sudden increase in $\sigma_m/\sigma_g$ for cobalt shown by the

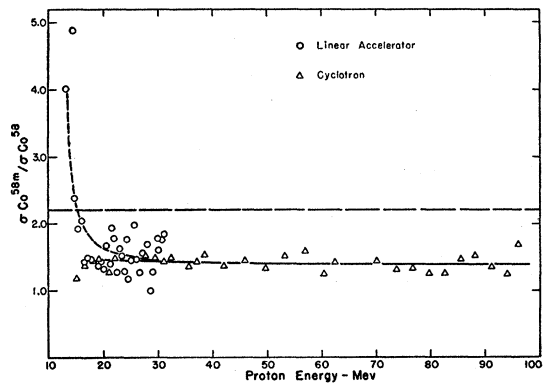


FIG. 6. $\sigma_m/\sigma_g$ for $Co^{58}$. The dashed line represents the ratio of the statistical weights.

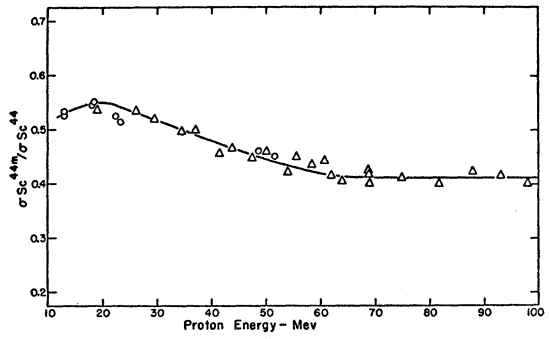


FIG. 7. $\sigma_m/\sigma_g$ for $Sc^{44}$. The ratio of statistical weights is 1.08–2.5.

linear accelerator data below 16 Mev (Fig. 6) is probably real, but the experimental error is very large due to the low counting rates. This effect would not be expected to appear in the cyclotron data. At the energies at which it occurs, the reaction cross section is very low and changing rapidly so it would be hidden by the poor energy resolution and the neutron background.

While this sudden increase in $\sigma_m/\sigma_g$ may indicate the breakdown of the applicability of statistical methods at low excitation energies, there is another possible explanation. The lowest energy studied (13 Mev) is only 1 Mev above the $(p,pn)$ threshold and 3.3 Mev above the $(p,d)$ threshold. Because of the small amount of energy available, particles must be emitted with very little orbital angular momentum and the gamma cascade will be very limited. Therefore large spin changes in the de-excitation process will be unlikely, and the compound nucleus might well be expected to decay to whatever isomeric state is closest to it in spin. Since the spin distribution of the compound nuclei will be approximately that given by the 10-Mev curve for $Sc^{45}$ in Fig. 8, a large relative yield of the high-spin isomer might be expected. If this explanation is correct, a similar experiment with good energy resolution should yield similar results for $Sc^{45}$ or other high-spin target nuclei.

At the energies of the cross-section maxima (20–30 Mev), the relative contribution of the compound nucleus mechanism should be at its greatest. With increasing energy the contribution of the knock-on mechanism should steadily increase until at energies near 100 Mev the reaction should proceed entirely by this mechanism. The relative yields of the high spin isomer for bromine and scandium (Figs. 5 and 7) begin to decrease at energies above that of the cross section maxima, finally leveling off at about 70 Mev and remaining constant from there on. The relative yield of the high-spin isomer for cobalt is constant to 100 Mev (Fig. 6). This would seem to indicate that the knock-on mechanism produces a distribution of spins in the residual nuclei which in general is lower than that produced by compound nucleus formation at proton energies of 20–30 Mev and which is independent of energy. There are two knock-on processes which can lead to a $(p,pn)$ reaction and which contribute about equally.[22] Both, in the limiting case of very high energy, are the result of a single quasi-elastic interaction with a nuclear particle. In the first case, one of the collision partners escapes without further interaction leaving the residual nucleus with <20-Mev excitation energy. Since the target nuclei have odd mass numbers, the maximum spin of the residual nucleus will be the sum of three single-particle states. In the second case, both collision partners escape, leaving the residual nucleus with an excitation energy less than the binding energy of the next nucleon. The maximum spin will then be the sum of two single-particle states. Thus the average value of the spin would be expected to be low and since there is only a limited range of excitation energy permitted, the distribution of spin should show little variation with bombarding energy.

The constant value of $\sigma_m/\sigma_g$ for the $Co^{59}(p,pn)Co^{58,58m}$ reaction is in great contrast to the results for the $Mn^{55}(\alpha,n)Co^{58,58m}$ reaction[8] which gave a rapid increase from ~1 at 10 Mev to 3.7 at 23 Mev, but it is not contradictory. $Mn^{55}$ has a spin of 5/2. Since alpha particles have much lower angular momentum barriers than protons, the compound nuclei formed with increasing alpha-particle energy will very rapidly include higher and higher spin states. The change in the distribution of $J$ should be somewhat similar to the case of protons on $Br^{81}$ although with a much higher average value. Thus a rapid increase of $\sigma_m/\sigma_g$ to a large value would be expected.

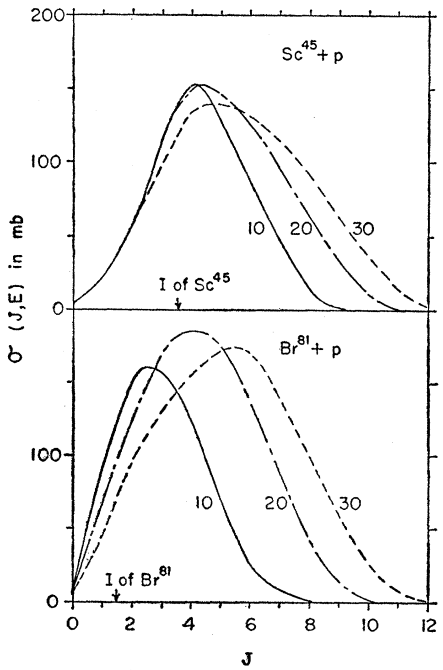


FIG. 8. Distribution of angular momentum in compound nuclei for proton energies of 10, 20, and 30 Mev.

[22] J. W. Meadows, Phys. Rev. 98, 744 (1955).

ACKNOWLEDGMENTS

The authors wish to thank Mr. S. Carpenter and Mr. M. Rice for their assistance in performing these experiments and Mr. Richard C. Wharton and the cyclotron crew for their cooperation in making the bombardments. One of the authors (R.M.D.) particularly wishes to acknowledge his helpful discussions with Professor P. Morrison.

PHYSICAL REVIEW VOLUME 102, NUMBER 1 APRIL 1, 1956

# Decay Scheme of $Au^{194}$†

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The radiations of the 40-hr $Au^{194}$ were investigated with a scintillation and a magnetic lens spectrometer, using coincidence techniques. First forbidden positron transitions with endpoints of 1.55±0.02 and 1.21 ±0.02 Mev were found to lead to the ground state and the first excited state of $Pt^{194}$. Most of the transitions and levels of $Pt^{194}$ known from the decay of $Ir^{194}$ were confirmed. Additional gamma rays of ~950, 1590±20, 1890±20, and 2150±15 kev were observed, indicating new levels at 2150 and 2215 kev.

## I. INTRODUCTION

THE decay of $Au^{194}$ to $Pt^{194}$ was first investigated in detail by Steffen *et al.*[1] who reported three gamma rays of 1480, 328, and 291 kev in cascade, with a cross-over transition of 2.1 Mev, and a weak 466-kev line whose position in the decay scheme was uncertain. The recent investigations of the gamma rays in $Pt^{194}$, following the beta decay of $Ir^{194}$, by Butement and Poë[2] (scintillation spectrometer), Johns and Nablo[3] (photoelectrons), and Mandeville *et al.*[4] (coincidence scintillation spectrometer) reveal a rather more complex level structure of $Pt^{194}$. The present work on the decay of $Au^{194}$ supplements these studies by additional coincidence measurements, by the approximate determination of conversion coefficients, and by the extension of the level scheme to higher energies.

## II. MEASUREMENTS

$Au^{194}$ was produced, together with several other Au isotopes, by bombarding Pt with the internal 9.5-Mev deuteron beam of the cyclotron. The method of source preparation and the apparatus used, a lens-type spectrometer with provision for coincidence counting, have been described elsewhere.[5,6]

### Gamma-Ray Spectrum

The pulse-height distribution from a NaI crystal spectrometer is shown in Fig. 1. As a guide to analyze it into its components, the pulse-height distributions from the 1.16-Mev gamma ray of $Sc^{44}$ and the 2.1-Mev gamma ray of $K^{38}$ were employed. The relative intensities of the components were then calculated by using the absorption coefficients of NaI and the photopeak

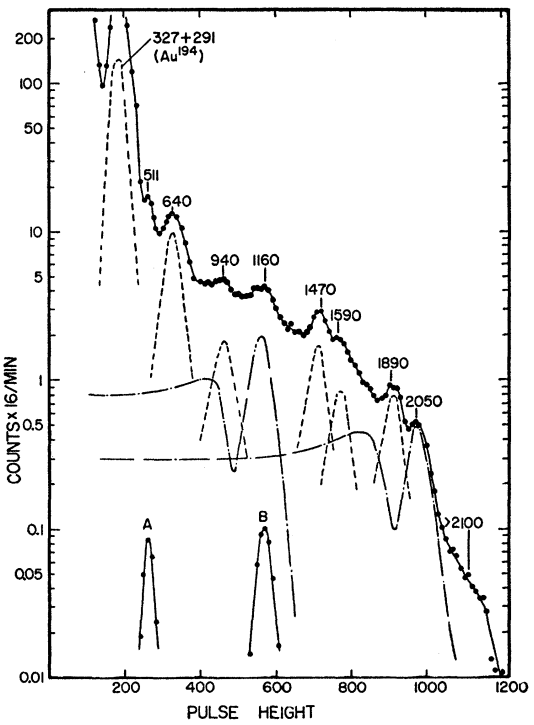


FIG. 1. Pulse-height spectrum (NaI) of the $Au^{194}$ gamma rays. The gross curve is analyzed into its components, whose full shape is indicated in two cases (1160 and 2050 kev), whereas only the photopeaks are shown for the other components. Curves *A* and *B* are the photopeaks of the annihilation radiation and the 1.16-Mev gamma ray of $Sc^{44}$, used for the energy calibration.

† Work supported by the U. S. Atomic Energy Commission. Part of a doctoral thesis submitted by M. T. Thieme to the faculty of Purdue University.
* Now at Los Alamos Scientific Laboratory.
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