

## Yield Ratios of Isomers Produced by Neutron Activation\*

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Values of the yield ratios of 14 pairs of isomers ( $\text{Co}^{60,60m}$ ,  $\text{Zn}^{69,69m}$ ,  $\text{Se}^{81,81m}$ ,  $\text{Br}^{80,80m}$ ,  $\text{Rb}^{86,86m}$ ,  $\text{Rh}^{104,104m}$ ,  $\text{Ag}^{110,110m}$ ,  $\text{In}^{114,114m}$ ,  $\text{Sb}^{122,122m}$ ,  $\text{Te}^{127,127m}$ ,  $\text{Cs}^{134,134m}$ ,  $\text{Ce}^{137,137m}$ ,  $\text{Eu}^{152,152m}$ , and  $\text{Ir}^{192,192m}$ ) produced by thermal neutron activation are given. The values are compared with those calculated by the method of Huizenga and Vandenbosch. From this comparison, the spins of several compound nuclei are indicated. Comparison of isomer yield ratios measured with thermal and with epi-cadmium neutrons are given for 7 pairs ( $\text{Rh}^{104,104m}$ ,  $\text{In}^{114,114m}$ ,  $\text{In}^{116,116m}$ ,  $\text{Cs}^{134,134m}$ ,  $\text{Eu}^{152,152m}$ ,  $\text{Ta}^{182,182m}$ , and  $\text{Ir}^{192,192m}$ ). These are discussed in terms of the spins of the compound nucleus which are associated with different neutron absorption resonances. New thermal neutron cross-section values are given for the formation of 9 nuclides ( $\text{Co}^{60m}$ ,  $\text{Rb}^{86m}$ ,  $\text{Rb}^{86}$ ,  $\text{Ag}^{110}$ ,  $\text{In}^{114m}$ ,  $\text{In}^{114}$ ,  $\text{Ir}^{192m}$ ,  $\text{Ir}^{192}$ , and  $\text{Ir}^{194}$ ).

### I. INTRODUCTION

THE relative yields of pairs of isomers produced in nuclear reactions have been studied by many workers in the quest for better understanding of the mechanism of such reactions. This paper is concerned with the production of isomers by activation with neutrons of low energy (thermal to a few eV). A qualitative rule,<sup>1</sup> that the isomer most favored is the one of spin nearest that of the compound nucleus (formed by addition of a neutron to a target nucleus), has been borne out by all the currently available data. Huizenga and Vandenbosch<sup>2</sup> have attempted to develop a scheme relating quantitative yield ratios to such physical quantities as the number of  $\gamma$  rays in cascade leading to the formation of the isomers, the spins of the isomers and of the compound nuclei, and a nuclear-spin density parameter. There has been, however, a scarcity of accurate values for isomeric yield ratios so that support or refutation of any theory has been difficult. Yield ratio, as used here, is defined as the ratio of the cross section for formation of the upper state to that of the lower state. It is the primary purpose of this paper to present new and more accurate values for several yield ratios. Also, where applicable, the effects of resonance neutrons on the measured yield ratios will be discussed. When large discrepancies were found between yield ratios and current values for cross sections, improved cross-section measurements were also made.

### II. EXPERIMENTAL

As defined above, a yield ratio may be obtained simply by dividing the cross section for the formation of the upper state by that of the lower state. Such a procedure applied to existing cross-section data leads to gross inaccuracies due to the fact that many of the available cross-section data are quite old and were ob-

tained by obsolete methods. Even the better recent direct determinations contain uncertainties arising from flux measurements and absolute disintegration rate determinations. Many of the older data were obtained without benefit of the present-day refinements in counting equipment, the availability of separated stable isotopes, and, in a few cases, without adequate knowledge of the decay schemes of the products. Whenever possible, the present measurements were made in such a way as to eliminate dependence on flux determination, decay scheme data, and number of target atoms. The following experimental descriptions explain these procedures.

For the isomers to be considered, three possible situations appear in regard to their decay schemes. The first type is that in which the upper state is longer lived than the lower and decays nearly entirely by isomeric transition. In this case the equations

$$A_m = N\varphi\sigma_m(1 - e^{-\lambda_m t}) \quad (1)$$

$$A_g = N\varphi\{(\sigma_m + \sigma_g)(1 - e^{-\lambda_g t}) + [\lambda_g\sigma_m/(\lambda_m - \lambda_g)](e^{-\lambda_m t} - e^{-\lambda_g t})\} \quad (2)$$

are applied. These are given in a slightly different form by Segrè and Helmholtz.<sup>3</sup>  $A_m$  and  $A_g$  are the activities of the upper and lower states at the time of removal from the reactor,  $N$  is the number of target atoms present during irradiation,  $\varphi$  is the flux,  $t$  is the irradiation time,  $\sigma_m$  and  $\sigma_g$  are the cross sections for the formation of the upper and lower states, and  $\lambda_m$  and  $\lambda_g$  are the decay constants of the upper and lower states. Often the equations may be simplified. Such simplification results if  $t \gg 1/\lambda_g$  or  $1/\lambda_m$ , or if either of the decay constants or cross sections is much larger than the other.

The second type is that in which the upper isomer is shorter lived than the lower and decays nearly entirely by isomeric transition. Here, Eq. (1) is used as is, but Eq. (2) is simplified (after the upper state has completely decayed), as

$$A_g' = N\varphi(\sigma_m + \sigma_g)(1 - e^{-\lambda_g t}), \quad (3)$$

<sup>3</sup>E. Segrè and A. C. Helmholtz, *Rev. Mod. Phys.* **21**, 271 (1949).

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<sup>1</sup>E. der Mateosian and M. Goldhaber, *Phys. Rev.* **108**, 766 (1957).

<sup>2</sup>J. R. Huizenga and R. Vandenbosch, *Phys. Rev.* **120**, 1305 (1960).

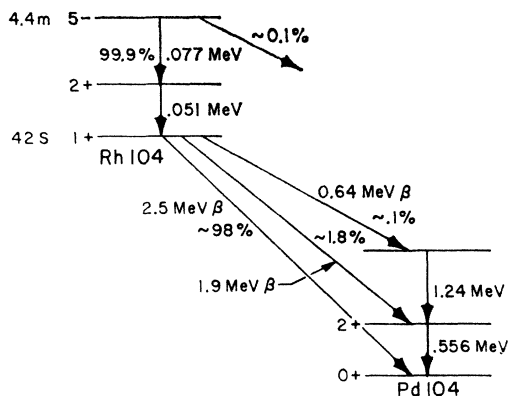
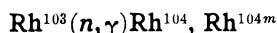


FIG. 1. Decay scheme of  $\text{Rh}^{104,104m}$ .

where  $A_p'$  is the measured activity of the lower state extrapolated back to the time of removal from the reactor.

The third type is that in which the two isomers decay almost entirely independently. Now the equation for calculating the formation cross section for the lower state becomes identical in form to Eq. (1).

All of the above are based on the assumption that there is no significant second-order capture during the irradiation. For this reason irradiations in the present work were kept as short and at as low a flux as was convenient.



The determination of the yield ratio of the  $\text{Rh}^{104}$  isomers is the best example of a "direct" measurement.

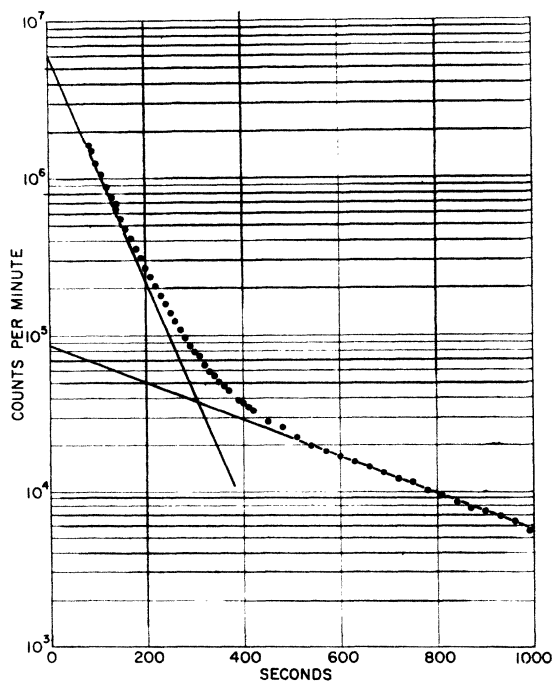
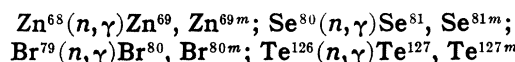
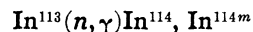


FIG. 2. Typical  $\text{Rh}^{104,104m}$  decay curve.

An examination of the decay scheme (Fig. 1) makes it obvious that by following the decay of the high energy  $\beta$  particles emitted by the lower isomer, one may determine the relative activities of the two states independently of the detector geometry and efficiency. Figure 2 shows a typical decay curve obtained by feeding discriminated, scaled pulses from an anthracene-crystal  $\beta$  detector into a fast strip-chart recorder. Extrapolation of the two portions of the curve gives the activities at the time of removal from the reactor. The target material, pure  $\text{Rh}^{103}$ , produces no other significant activities. Since only one sample is used for the determination of both isomers, the yield ratio is independent of flux and target weight. The only sources of error in this determination, therefore, are in timing and in counting statistics. These experiments gave consistently reproducible results to within less than 2%. The yield ratio for the rhodium isomers was determined for thermal neutrons, for epi-cadmium neutrons, and for 1.1-eV neutrons (roughly corresponding to the resonance in rhodium which peaks at 1.2 eV). The latter monoenergetic neutrons were obtained with the MTR crystal spectrometer.



The yield ratios of the  $\text{Zn}^{69}$ ,  $\text{Se}^{81}$ ,  $\text{Br}^{80}$ , and  $\text{Te}^{127}$  isomers were determined in the same way as in the case of rhodium but with less accuracy due to poorer counting statistics, and/or some interference from other activities. These were largely eliminated in the case of Se and Te by using 98 and 94.5% enriched  $\text{Se}^{80}$  and  $\text{Te}^{126}$ , respectively. The yield ratios for this group were determined only with thermal neutrons because the target isotopes do not have any resonances in the region of interest.



For the determination of the yield ratio of the  $\text{In}^{114}$  isomers, the same procedure as that used for rhodium was used with some modifications. The decay was followed with an end-window  $\beta$  proportional counter (using an absorber to eliminate any conversion electrons) and the data were recorded with an automatic data printing system ordinarily used with a  $4\pi\beta$  counter.<sup>4</sup> However, due to a large difference in activities of the two isomers produced, a reirradiation of the sample was necessary (after the activity of the short-lived isomer was determined) for 100 times as long as the original irradiation. This necessitated the use of flux monitors for the two irradiations, but by using the same material as a monitor and by re-irradiating the sample in the same position in the reactor, errors in the relative flux determination were minimized. Enriched

<sup>4</sup> B. Keisch, Quarterly Report, MTR-ETR Technical Branch, Atomic Energy Commission Report IDO-16733, December, 1961 (unpublished), p. 13.

96%  $\text{In}^{113}$  was used in order to partially eliminate the interference of the 54-min  $\text{In}^{116}$  activity produced. The cross sections for the formation of the isomers were also measured because the yield ratio was in serious disagreement with the data in the literature (see results). For the necessary absolute-activity determinations, the counter arrangement described above was calibrated using a  $\text{Re}^{188}$  source (which emits  $\beta$  particles of essentially the same energy as  $\text{In}^{114}$ ). The  $\text{Re}^{188}$  source was prepared by irradiating 98.8% enriched  $\text{Re}^{187}$ , standardized with the  $4\pi\beta\text{-}\gamma$  coincidence system and afterwards mounted intact in the same way as the  $\text{In}^{114}$  sources. Gold-foil monitors were used for the neutron flux determinations. Yield ratios for the  $\text{In}^{114}$  isomers were determined for both thermal and epi-cadmium neutrons.

#### $\text{Eu}^{151}(n,\gamma)\text{Eu}^{152}, \text{Eu}^{152m}$

The yield ratio for the  $\text{Eu}^{152}$  isomers could not be determined by counting the same radiation for each isomer since these isomers decay independently. The method chosen was based on counting the 344- and 122-keV  $\gamma$  rays which are both emitted (but with different branching ratios) in the decay of each isomer, and averaging the results. Thus, an error due to uncertainties in the branching ratios was introduced. The material used was 92.1% enriched  $\text{Eu}^{151}$  which eliminated most of the interference from the  $\text{Eu}^{154}$  also produced. A measurement was also made of the ratio of the yield ratio with thermal neutrons to that with epi-cadmium neutrons. This ratio of ratios is independent of errors in the decay scheme because the measurements were made in the same way for the two neutron energy ranges.

#### $\text{Ce}^{136}(n,\gamma)\text{Ce}^{137}, \text{Ce}^{137m}$

The decay of the  $\text{Ce}^{137}$  isomers was followed as for the case of the rhodium isomers but with the following exceptions. The radiation detected was the 440-keV  $\gamma$  ray which occurs in 3% of the disintegrations of the lower isomer. This was done by using a single-channel analyzer with the window set to include this peak only. Chemical purification from other rare earths and zirconium was necessary before counting, and this was done either by ion exchange, or by a reduction-oxidation-precipitation cycle. The interference from the  $\text{Ce}^{143}$  activity found to be present was first reduced by using 30% enriched  $\text{Ce}^{136}$ . A correction was then applied to the data based on fitting the  $\gamma$ -ray spectrum of pure  $\text{Ce}^{143}$  to that of the sample and subtracting. The yield ratio in this case was determined for thermal neutrons only.

#### $\text{Rb}^{85}(n,\gamma)\text{Rb}^{86}, \text{Rb}^{86m}$

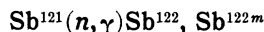
A  $\gamma$ -ray scintillation detector connected to a 256-channel analyzer was used to determine the yield ratio for the  $\text{Rb}^{86}$  isomers. The upper state decays by isomeric transition with a 1.02-min half-life, emitting a 560-keV

$\gamma$  ray. A sample was prepared with an activity chosen to give a small, correctable dead time and the 1-min activity was "counted to death" on the multichannel analyzer. The number of  $\gamma$  rays (corrected for geometry, efficiency, peak-to-total ratio, and absorption) represented by the 560-keV peak then represents the number of  $\text{Rb}^{86m}$  nuclei present at the start of the count and is easily converted to activity. After a period long enough for the 18-min  $\text{Rb}^{88}$  activity present to decay, another count was taken on the same sample in order to determine the activity of the 19-day  $\text{Rb}^{86}$ . This was done by counting the 1080-keV  $\gamma$  ray representing 8.9% of the transitions in its decay. Since the yield ratio disagreed considerably with the cross-section data in the current literature (see results), the cross sections for formation of the two isomers were separately determined. For this purpose, the counting technique described above was used for measurement of the activity of the 1-min isomer. Then  $4\pi\beta\text{-}\gamma$  coincidence counting was used for determination of the 19-day activity (with decay followed as a purity check). Gold-foil monitors were used for neutron flux measurements. Only thermal neutrons were used for the determination of these values.

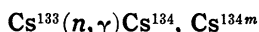
#### $\text{Ir}^{191}(n,\gamma)\text{Ir}^{192}, \text{Ir}^{192m}$

In this case it is necessary to determine the absolute activities of the two isomers because the upper isomer is shorter-lived. However, it is possible to determine the yield ratio with only one sample so that flux and target-atom measurements are not necessary. The upper state (1.45-min half-life) decays essentially entirely by a highly converted 57-keV isomeric transition to the lower state. The conversion electrons from this process as well as the  $\beta$  particle from the decay of the 74.5-day isomer were counted in a special  $4\pi\beta$  counter having a beryllium window so that the conversion x rays ( $\sim 12$  keV) could be detected by the  $\gamma$  coincidence apparatus. This enables one to measure the efficiency of the  $\beta$  detector for the electrons. The decay of the 1.45-min activity was followed automatically as was the 19-h  $\text{Ir}^{194}$  activity also present. (The latter activity was also determined.) Due to the fact that for a given iridium sample, the activities of the two isomers are very different in intensity, only a separate small part of any one sample was counted for the 1.45-min activity and the two portions resulting were inter-compared by means of the 19-h  $\text{Ir}^{194}$  activity in each portion. Excellent reproducibility among separate runs was obtained in this way. A gold-foil flux monitor was included in each irradiation and the irradiated samples were weighed so that cross sections for the formation of the three ( $\text{Ir}^{192}, \text{Ir}^{192m}, \text{Ir}^{194}$ ) radioactive nuclides produced could also be determined. The relative yield ratios for thermal and epi-cadmium neutrons were measured by counting the unconverted 57-keV  $\gamma$  rays from the 1.45-min isomer and the 317-keV  $\gamma$  rays from the 74.5-day isomer in a fixed geometry for all samples.

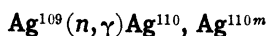
In addition, other measurements were made with both cadmium and silver covers in order to separate the effects of the 0.64- and 5.4-eV absorption resonances in Ir<sup>191</sup>. The latter corresponds closely in energy to that of a large resonance in Ag<sup>109</sup>.



A technique similar to that described above for the iridium isomers was used to determine the yield ratio of the Sb<sup>122</sup> isomers. In this case 99.4% enriched Sb<sup>121</sup> was used to eliminate interference from other antimony activities and the activities of both the short- and the long-lived isomers were measured on a single sample. The decay was followed with the 4 $\pi$  $\beta$  counter for a time sufficient to evaluate the effect of a slight amount of unidentified impurity detected in the sample. Efficiencies were determined by  $\gamma$ -coincidence counting at appropriate intervals. The half-life of the shorter-lived isomer as given in the current literature was found to be erroneous (see results). It was, therefore, carefully measured by following the decay (over several half-lives) of the unconverted 75- and 61-keV  $\gamma$  rays of the isomeric transition with a single-channel analyzer and the automatic counting equipment. The results were analyzed by the IBM 650 computer making a weighted least-squares fit of the data. No attempt was made to measure the yield ratio with anything but thermal neutrons in this case.

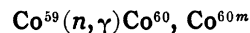


Here the appearance of recent cross-section data in the literature<sup>5,6</sup> and certain difficulties in a direct determination make it more feasible to calculate the yield ratio from the cross sections. However, the ratio of the yield ratios for thermal and epi-cadmium neutrons was easily determined by the same method ( $\gamma$ -ray counting in a fixed geometry) as that used with the iridium isomers.

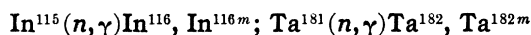


Here again it is very difficult to make a direct measurement of the yield ratio. In this case, it was felt that some improvement could be made in the cross-section value for the formation of 24-sec Ag<sup>110</sup> by a new determination. The literature value was suspected to be in error since it is larger than the accepted value of the absorption cross section (see results). A new value for the formation cross section was determined by irradiating thinly deposited weighed samples of AgNO<sub>3</sub> which had been sealed between two thin polyethylene films, and counting the  $\beta$  activity "to death" in a 4 $\pi$  $\beta$  counter. The efficiency for the detector was determined during the counts and was about 90–92% for the samples. A gold-foil monitor was used for the neutron flux measure-

ments. This determination was made for thermal neutrons only.



For the determination of the yield ratio of the Co<sup>60</sup> isomers, it was decided that the best method would be to remeasure the cross section for the formation of the 10.5-min upper state. This was done by counting the unconverted 59-keV  $\gamma$  rays from the Co<sup>60m</sup> produced, and applying a correction based on a fairly well-known conversion coefficient.<sup>7</sup> A cobalt monitor was used for flux measurement so that some of the errors in this determination were minimized when the yield ratio was calculated. This determination was done only for thermal neutrons.



In these cases it was impossible to determine an accurate yield ratio directly due to a short half-life in the case of In<sup>116m</sup> and due to uncertainties in the decay scheme in the case of Ta<sup>182m</sup>. However, relative yield ratios for thermal and epi-cadmium neutrons are of interest due to the high resonance absorption in In<sup>115</sup> and Ta<sup>181</sup>. These relative yield ratios were determined by  $\beta$ -particle counting with fixed geometries.

### III. RESULTS AND DISCUSSION

#### A. The Thermal Neutron Yield Ratios

The yield ratios obtained with thermal neutrons are given in Table I. Also given are the spins of the target and product nuclei and the yield ratios calculated by the method of Huizenga and Vandenbosch.<sup>2</sup>

Their scheme involves the assumption that, after the formation of a compound nucleus by capture of an *s*-wave neutron, a series of electric dipole  $\gamma$  rays is emitted in cascade until the isomeric states are reached. The number of  $\gamma$  rays in such a cascade is assumed to be 3 to 5 based on observed  $\gamma$ -ray multiplicities, and for each step it is assumed that many spin states are available for population. The probable distribution of spins after each  $\gamma$  ray, is calculated assuming the distribution is proportional to

$$(2J+1)e^{-(J+1/2)^2}/2\sigma^2, \quad (4)$$

where *J* is the resulting spin and  $\sigma$  is a spin distribution parameter. This parameter is varied from 3 to 5 which is typical of observed values. The calculated distribution, after the emission of the next-to-the-last  $\gamma$  ray, is used to determine the relative amounts of the isomers produced, by assuming that those states having spins closest to one of the isomers will go to that isomer. The statistical treatment becomes less valid where many one- or two-step transitions to the isomers occur as in the lighter nuclides.<sup>2</sup>

<sup>5</sup> B. Keisch, *J. Inorg. & Nucl. Chem.* **17**, 180 (1961).

<sup>6</sup> F. Brown, P. J. Campion, and B. H. Oliver, *J. Nucl. Energy, Pt. A, Reactor Sci.* **13**, 141-4 (1961).

<sup>7</sup> J. H. Kahn, Oak Ridge National Laboratory Report, ORNL-1089, 1951 (unpublished).

TABLE I. Thermal neutron yield ratios.

Target nucleus	Target spin (I)	Upper state spin	Lower state spin <sup>a</sup>	Experimental thermal neutron yield ratio	Compound nucleus spin ( $J_c$ )	Yield ratios calculated by method of Huizenga and Vandenbosch (reference 2)					
						$N_\gamma=3$ $\sigma=3$	$N_\gamma=4$ $\sigma=3$	$N_\gamma=5$ $\sigma=3$	$N_\gamma=3$ $\sigma=5$	$N_\gamma=4$ $\sigma=5$	$N_\gamma=5$ $\sigma=5$
Co <sup>68b</sup>	7/2-	2	5	1.19 ±0.16	$I-\frac{1}{2}$ $I+\frac{1}{2}$	2.66 0.776	2.35 0.996	2.31 1.11	1.57 0.435	1.26 0.497	1.15 0.508
Zn <sup>68b</sup>	0+	9/2	1/2	0.100±0.005	$\frac{1}{2}$	0.126	0.259	0.587	0.181	0.395	0.515
Se <sup>80b</sup>	0+	7/2	1/2	0.21 ±0.01	$\frac{1}{2}$	0.355	0.593	0.848	0.443	0.792	1.20
Br <sup>79b</sup>	3/2-	5	1(2)	0.31 ±0.02	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0 0.119	0.055 0.161	0.095 0.203	0 0.193	0.095 0.291	0.175 0.406
Rb <sup>85</sup>	5/2-	6	2	0.12 ±0.01	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0.056 0.208	0.089 0.239	0.121 0.256	0.088 0.350	0.163 0.445	0.236 0.520
Rh <sup>103</sup>	1/2-	5	1(2)	0.075±0.002	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0 0	0 0.055	0.055 0.095	0 0	0 0.095	0.095 0.175
Ag <sup>109</sup>	1/2-	6	1(2) <sup>a</sup>	0.036±0.005	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0 0	0 0.027	0.027 0.053	0 0	0 0.045	0.045 0.099
In <sup>113</sup>	9/2+	5	1	2.1 ±0.1	$I-\frac{1}{2}$ $I+\frac{1}{2}$	2.43 9.21	1.88 4.88	1.58 3.34	4.13 17.7	3.52 10.8	3.25 8.26
Sb <sup>121</sup>	5/2+	7	2(4)	0.066±0.008	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0 0	0 0.015	0.005 0.025	0 0	0 0.021	0.019 0.073
Te <sup>126</sup>	0+	11/2	3/2	0.15 ±0.02	$\frac{1}{2}$	0	0.045	0.093	0	0.066	0.147
Cs <sup>133</sup>	7/2+	8	4(5)	0.079±0.008	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0 0	0 0.008	0.003 0.011	0 0	0 0.033	0.012 0.056
Ce <sup>136</sup>	0+	11/2	3/2	0.15 ±0.01	$\frac{1}{2}$	0	0.045	0.093	0	0.066	0.147
Eu <sup>151</sup>	5/2+	0	3	0.56 ±0.04	$I-\frac{1}{2}$ $I+\frac{1}{2}$	0.31 0.11	0.31 0.13	0.30 0.16	0.22 0.067	0.22 0.079	0.19 0.090
Ir <sup>191</sup>	3/2+	1 <sup>a</sup>	4	2.0 ±0.1	$I-\frac{1}{2}$ $I+\frac{1}{2}$	4.6 1.7	3.1 1.4	2.2 1.3	3.4 1.2	2.1 0.93	1.4 0.76

<sup>a</sup> All spins taken from *Nuclear Data Sheets*, National Academy of Science (National Research Council, Washington, D. C.), except Ag<sup>109</sup>, taken from N. M. Anton'eva, A. A. Bashilov, and E. A. Kulakovskii, *Soviet Phys.—JETP* 10, 1063 (1960) and I<sup>191</sup>, taken from reference 10. Spins in parenthesis indicate a state intermediate between upper and lower isomers which would feed the lower isomer.

<sup>b</sup> Statistical model probably not applicable.

Examination of Table I will disclose that for most of the isomers, a reasonable choice of the number of  $\gamma$ 's ( $N_\gamma$ ) in the cascade and of  $\sigma$  will give a result agreeing with the experimental value. *If this model is assumed correct*, in some cases a choice exists which enables one to determine, to some extent, the values of  $N_\gamma$ ,  $\sigma$ , and/or  $J_c$  (as  $I \pm \frac{1}{2}$ ). For example, in the case of In<sup>113</sup>,  $J_c = I - \frac{1}{2}$  and  $\sigma$  is definitely 3 or 4. In the case of Sb<sup>121</sup>,  $J_c = I + \frac{1}{2}$  and  $\sigma$  and  $N_\gamma$  are large. Note also for Te<sup>126</sup> and Ce<sup>136</sup> the similarity of spins and of yield ratios and the fact that both cases require  $N_\gamma = 5$  and  $\sigma = 5$ . In the cases of Br<sup>79</sup>, Rb<sup>85</sup>, In<sup>113</sup>, Sb<sup>121</sup>, Cs<sup>133</sup>, Eu<sup>151</sup>, and Ir<sup>191</sup>, there are strong indications of the value of  $J_c$ . For others, notably Co<sup>69</sup>, Rh<sup>103</sup>, and Ag<sup>109</sup>,  $J_c$  is not determinable from this model. Some of the data show poor agreement between the calculated and experimental results no matter what choices of  $N_\gamma$  and  $\sigma$  are made.

For Zn<sup>68</sup> and Se<sup>80</sup>, somewhat lower values of the parameters are indicated. As has been pointed out by Bishop,<sup>8</sup>  $\sigma$  is dependent on  $A^{7/12}$  and so for these light nuclei a choice of a lower value for  $\sigma$  is possible. Also for the lighter targets, many high energy  $\gamma$  rays have been observed<sup>9</sup> indicating a smaller value for  $N_\gamma$  and

that the statistical model is probably not applicable. Bishop obtained a still lower value for the Se<sup>80</sup> yield ratio with thermal neutrons but approximately the same value for epi-cadmium neutrons as is presented here for the thermal case.

For Cs<sup>133</sup> it would appear that a greater value of  $N_\gamma$  or  $\sigma$  would be required. For example,  $N_\gamma = 6$ ,  $\sigma = 5$  gives 0.080 for the calculated value of the yield ratio which agrees with the experimental value.

For Eu<sup>151</sup>, a low value for  $N_\gamma$  and/or  $\sigma$  would be required. Here, however, there exists the possibility that the experimental yield ratio may be wrong since it does depend on the accuracy of two rather complicated decay schemes.

The value of the yield ratio for the Ir<sup>192</sup> isomers warrants some separate discussion. Scharff-Goldhaber and McKeown<sup>10</sup> obtained a value of 0.84 by studying the growth of the intensity of a  $\gamma$  ray (468 keV) from a state presumably not populated in the decay of Ir<sup>192m</sup>. Although such a value ( $< 1$ ) does not agree with the qualitative<sup>1</sup> isomer ratio rule relating compound nucleus spin and isomer spins, it would appear that the model of Huizenga and Vandenbosch does allow such a value. That is, with  $J_c = I + \frac{1}{2}$ ,  $\sigma = 5$ , and  $N_\gamma = 4$  or 5, such a value is calculated. However, this measurement is difficult to perform and relatively insensitive. An at-

<sup>8</sup> C. T. Bishop, Argonne National Laboratory Report ANL-6405, 1961 (unpublished).

<sup>9</sup> L. V. Groshev, A. M. Demidov, V. N. Lutsenko, and V. I. Pelekhov, *Atlas of  $\gamma$ -ray Spectra from Radiative Capture of Thermal Neutrons*, translated from the Russian by J. B. Sykes (Pergamon Press, New York, 1959).

<sup>10</sup> G. Scharff-Goldhaber and M. McKeown, *Phys. Rev. Letters* 3, 47, (1959).

TABLE II. New activation cross sections.

Product	Formation cross-sections thermal neutrons (b)
Co <sup>60m</sup>	20±2
Rb <sup>86m</sup>	0.061±0.003
Rb <sup>86</sup>	0.45 ±0.04
Ag <sup>110</sup>	89±4
In <sup>114m</sup>	8.1 ±0.8
In <sup>114</sup>	3.9 ±0.4
Ir <sup>192m</sup>	610±60
Ir <sup>192</sup>	300±30
Ir <sup>194</sup>	97±10

tempt by the present writer to duplicate Scharff-Goldhaber and McKeown's procedure gave the result  $Y=1.3\pm 0.5$ . Therefore, the method described above in the experimental section was developed, which gave a value of  $2.0\pm 0.1$ .

Finally, it is interesting to note that although the measured yield ratios all follow the qualitative isomer yield ratio rule,<sup>1</sup> the present model (as already indicated above in the case of Ir<sup>191</sup>) does allow some violation of this rule.

### B. Some New Cross-Section Values

Table II lists some new values for thermal neutron formation cross sections which were incidentally obtained as indicated in the experimental section. All neutron flux determinations were made using appropriate cross sections and half-lives as given by Hughes and Schwartz.<sup>11</sup>

The cross section for formation of Co<sup>60m</sup> agrees well with that obtained by Moss and Yaffee<sup>12</sup> by a different method ( $18.3\pm 1.7$  b) and not with that given in various compilations<sup>11,13</sup> (16 b).

The value given in the table for the formation of Ag<sup>110</sup> agrees quite well with the absorption cross section of Ag<sup>109</sup> ( $85\pm 8$  b) in the literature<sup>11</sup> while the presently accepted value (113 b) does not.<sup>11</sup>

It has been noted throughout this work that large errors in the published values of cross sections for isomer formation were frequently encountered. In many cases improved values may now be obtained using better equipment, materials, and decay-scheme knowledge than were previously available.

Values for the other nuclides in Table II represent similar changes from the literature values presently in use.<sup>11,13</sup>

### C. The Half-Life of Sb<sup>122m</sup>

As mentioned in the experimental section a re-determination of the half-life of Sb<sup>122m</sup> was made giving

<sup>11</sup> D. J. Hughes and R. B. Schwartz, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (U. S. Government Printing Office, Washington, D. C., 1958); and Supplement No. 1, 1960.

<sup>12</sup> N. Moss and L. Yaffee, *Can. J. Chem.* **31**, 391 (1953).

<sup>13</sup> W. H. Sullivan, *Triblinear Chart of Nuclides* (U. S. Government Printing Office, Washington, D. C., 1961).

a value of  $4.21\pm 0.02$  min where the error quoted is at the 90% confidence limit. This compares with the previously accepted value of 3.5 min.<sup>11</sup>

### D. Yield Ratios with Epi-Cadmium Neutrons

Table III gives the variation of yield ratio with energy of the neutrons. It also shows the contribution of the resonances to the thermal neutron cross section in each case based on Breit-Wigner calculations.<sup>11</sup>

For the Rh<sup>104</sup> isomers, a very slight change (increase) in the yield ratio is noted for epi-cadmium neutrons versus thermal neutrons. The yield ratio determined with 1.1-eV neutrons (from the MTR crystal spectrometer) also shows this increase. In view of the fact that only a very small part of the thermal neutron cross section is due to the negative energy resonance, the definite, but small, change in the yield ratio probably indicates a shift in  $J_c$  (the spin of the compound nucleus). An examination of the calculated values in Table I indicates that the negative-energy resonance might be associated with  $J_c=I-\frac{1}{2}$  while the 1.257-eV resonance might be associated with  $J_c=I+\frac{1}{2}$ .

For the In<sup>114</sup> isomers, a similar argument can be made, but the  $Y_{Cd}/Y_{th}$  value is almost unity within experimental error. This would indicate that the spin of the compound nucleus has the same value for the negative-energy and the lower positive-energy resonance, namely,  $J_c=I-\frac{1}{2}$ .

Similar arguments applied to the other nuclides in Table III lead to the following conclusions:

In<sup>115</sup>—No difference in spin between the negative-energy and the 1.486-eV resonance is indicated or else any change is masked by the large contribution which the resonance makes to the thermal cross section. See Domanic and Sailor<sup>14</sup> and Stolovy<sup>15</sup> for discussion of other resonances.

Cs<sup>133</sup>—Probably  $J_c=I+\frac{1}{2}$  is associated with the negative-energy resonance and  $J_c=I-\frac{1}{2}$  with the 5.90-eV resonance.

Eu<sup>151</sup>—There is a slight indication that  $J_c=I+\frac{1}{2}$  is associated with the negative-energy resonance and  $J_c=I-\frac{1}{2}$  with one or more of the positive-energy resonances in the 0.3- to 1.1-eV range. See reference 2 for discussion of these resonances.

Ta<sup>181</sup>—A difference in spin exists for negative- and positive-energy resonances. Based on calculations of Huizenga and Vandenbosch,<sup>2</sup>  $J_c=I+\frac{1}{2}$  is associated with the negative-energy resonance and  $J_c=I-\frac{1}{2}$  with the 4.28-eV resonance.

Ir<sup>191</sup>—Apparently,  $J_c=I-\frac{1}{2}$  is associated with the negative-energy resonance and  $J_c=I+\frac{1}{2}$  with both the 0.654- and 5.36-eV resonances. This case is the best example given here because both  $Y_{th}=2.0$  and  $Y_{Cd}=0.68\times 2.0=1.4$  fit the model and with the same

<sup>14</sup> F. Domanic and V. L. Sailor, *Phys. Rev.* **119**, 208 (1960).

<sup>15</sup> A. Stolovy, *Phys. Rev.* **118**, 211 (1960).

TABLE III. Epi-cadmium-thermal yield ratios.

Target nucleus	% of thermal cross section due to positive resonance(s) <sup>a</sup>	Thermal neutron yield ratio ( $Y_{th}$ )	Epi-cadmium neutron yield ratio ( $Y_{Cd}$ )	$Y_{Cd}/Y_{th}$	Additional data
Rh <sup>103</sup>	99	0.075±0.002	0.080±0.001	1.07±0.03	With spectrometer neutrons at 1.1 eV $Y=0.080$ ±0.001
In <sup>113</sup>	36	2.1 ±0.1	2.4 ±0.2	1.14±0.12	
In <sup>115</sup>	95	...	...	1.01±0.06	$Y_{Cd}/Y_{Cd+Ag}$ = 1.00±0.05
Cs <sup>133</sup>	49	0.079±0.008	...	0.82±0.04	
Eu <sup>151</sup>	4,22	0.56 ±0.04	...	0.97±0.01	
Ta <sup>181</sup>	51	...	...	0.92±0.04	
Ir <sup>191</sup>	24,2	2.0 ±0.1	...	0.68±0.01	

<sup>a</sup> Contribution calculated for first positive resonance only except for Eu<sup>151</sup> and Ir<sup>191</sup> where the contribution of the first two are given in order.

$N_\gamma$  and  $\sigma$  values. The conclusions for the 5.36-eV resonance is not as certain as that for the 0.654-eV resonance because at best a very small effect with the silver cover would be observed. This is due to the much lower intensity of 5.4-eV neutrons in the reactor spectrum.

Resonance spin assignments have been made by other methods for several of the nuclides considered here. However, no independent spin assignments are yet available for comparison with any of the assignments made in the present work.<sup>16</sup>

#### D. Conclusions

In most cases, the scheme of Huizenga and Vandenberg turns out to be useful for explaining isotope yield ratios quantitatively. However, more specific information about the  $\gamma$  cascade following capture of a neutron

<sup>16</sup> B. A. Magurno and J. R. Stehn (private communication).

would be helpful, particularly for those nuclei which do not follow the typical pattern. In this connection, a discussion of certain aspects of this type of analysis is given by Huizenga and Vandenberg<sup>2</sup> and also by Bishop.<sup>8</sup> Determination of the compound nucleus spin is made somewhat more definite using this quantitative scheme than with the qualitative isomer ratio rule. Furthermore, in some cases, the possibility of ambiguities is brought out more clearly.

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