# Dependence of the $In^{116}$ Activation Ratio on Neutron Energy*

FAHRI DOMANIC† AND VANCE L. SAILOR
*Brookhaven National Laboratory, Upton, New York*
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Indium was irradiated with monochromatic neutrons of various energies, and a measurement was made of the ratio of the 54-minute to the 13-second activities of $In^{116}$ produced by neutron capture in $In^{115}$. Such a ratio expresses the relative probability for populating the ground or the isomeric state from the initial compound state. The irradiations were made with monochromatic neutrons from a crystal spectrometer at the resonance energies 1.456 ev and 3.86 ev, in regions between resonances at 0.1 and 2.66 ev, and with "pile neutrons." The results show that the activation ratio differs for the two resonances by a factor of approximately 3.5, with relatively more of the 13-second activity being associated with the 3.86-ev resonance. The half-lives of the two $In^{116}$ activities were redetermined and the values 13.4±0.4 seconds and 53.9±0.2 minutes were obtained.

## I. INTRODUCTION

SEVERAL years ago, prior to the general availability of enriched isotopes, the isotopic assignments of the slow neutron resonances in indium were made by activating indium foils with monochromatic neutrons and determining which resonances yielded $In^{116}$ activities.[1] In the course of this work it was found that both 1.458- and 3.86-ev resonances were in $In^{115}$, but it appeared that the ratio of the 54-minute to the 13-second $In^{116}$ activity was different for the two resonances. A similar situation was observed by Wood[2] in the isomeric activities of $Eu^{152}$. If such differences are indeed real, they are related to the gamma-ray cascades which follow neutron capture. Even though the compound nucleus is formed in a state of very high excitation and high spin ($E=6.38$ Mev, and $J=4$ or 5 for $In^{116}$), it is possible for the subsequent cascades from two neighboring compound states to terminate preferentially in two quite different states, i.e., the ground state or the metastable state. Direct experimental evidence for a difference in the cascade gamma rays at the two resonances has been found by Draper et al.[3]

Considerable information about the first two indium resonances is available. Landon[4] found that the radiation widths of these two resonances are significantly different. Even though the observed difference is only about 10%, such a fluctuation in $\Gamma_\gamma$ is highly improbable in terms of the distribution anticipated by Porter and Thomas[5] who show that the radiation widths should probably fit a chi-squared distribution with a large number of degrees of freedom. Such a distribution implies highly uniform radiation widths. A possible explanation for Landon's results is that the two resonances are in opposite spin states and that each spin state is characterized by a separate and independent Porter-Thomas distribution. The angular momenta of the compound states which correspond to $s$-wave resonances are limited to $J=I\pm\frac{1}{2}$, where $I$ is the spin of the target nucleus. The 1.458-ev resonance has the value $J=I+\frac{1}{2}=5$, as determined by the scattering measurements of Moore[6] and the nuclear polarization experiment of Dabbs et al.[7] Very recent nuclear polarization measurements by Stolovy[8] have confirmed the above assignment, and have yielded the value $J=I-\frac{1}{2}=4$ for the 3.86-ev resonance. Thus the spin assignments are not inconsistent with the fluctuations in $\Gamma_\gamma$ since the resonances do indeed appear to be in different spin states.

In view of the many interesting and basic features of these related phenomena it appeared desirable to confirm the previously observed differences in the activation ratio of $In^{116}$, particularly since the earlier measurements had been rather crude.

## II. EXPERIMENTAL DETAILS

When $In^{115}$ captures a neutron, the compound nucleus $In^{116}$ is formed in a well-defined excited state corresponding to one of the resonances in the neutron cross section. The compound state decays subsequently by emission of one or more capture gamma rays terminating in either the ground state of $In^{116}$ or an isomeric state at about 70-kev excitation. Both of these states decay to $Sn^{116}$ by $\beta$ emission with distinctly different half-lives (see Fig. 1); i.e., $T_{\frac{1}{2}}$(ground state) $=13.4$ seconds, $T_{\frac{1}{2}}(70\text{ kev})=53.9$ minutes. By measuring the saturated activities for these two half-lives we can determine the ratio for populating the ground and isomeric states from the initial compound state.

The indium foils were irradiated on a high intensity, low-resolution (1.6 μsec/m) crystal spectrometer using Be(10$\bar{1}$1) as monochromator. The foils were mounted

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† ICA Fellow, on leave from the University of Ankara, Ankara, Turkey.
[1] V. L. Sailor, Bull. Am. Phys. Soc. **27**, 46 (1952).
[2] R. E. Wood, Phys. Rev. **95**, 453 (1954).
[3] J. E. Draper, C. A. Fenstermacher, and H. L. Schultz, Phys. Rev. **111**, 906 (1958).
[4] H. H. Landon and V. L. Sailor, Phys. Rev. **98**, 1267 (1955).
[5] C. E. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).
[6] J. A. Moore, Phys. Rev. **109**, 417 (1958).
[7] J. W. T. Dabbs, L. D. Roberts, and S. Bernstein, Phys. Rev. **98**, 1512 (1955).
[8] A. Stolovy, Phys. Rev. **118**, 211 (1960).

on a sliding frame which permitted them to be moved quickly and precisely from the irradiation position to the counting position. Thus counting could begin a fraction of a second after the irradiation had ended. It was necessary, of course, to shield the foils against stray neutrons which copiously populate the experimental area, and to shield the proportional counters against environmental gamma-ray background with a heavy lead shield.

The radiations in the two activities are not identical. As can be seen from Fig. 1, the beta particles emitted in the 13-second decay are much more energetic than in the 54 minute. The 54-minute beta activity is accompanied by various gamma rays, while the 13 second is pure beta decay. In order to count the two activities with approximately equal efficiency we have used two end window gas flow counters having window thicknesses of only 0.9 mg/cm². The two counters

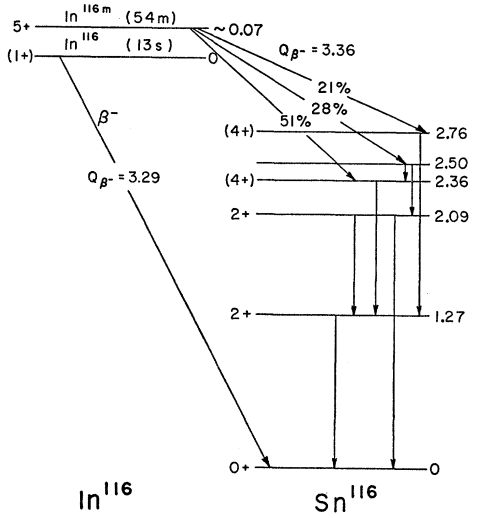


FIG. 1. The decay scheme of $In^{116}$ (taken from reference 10).

were placed on each side of the foil to approach $4\pi$ counting geometry, the separation being 8 mm and the window diameter being 1 inch. The counters were operated in the proportional region each with a cathode-follower preamplifier. The output of the preamplifiers were connected in parallel to a linear amplifier and then to the scaler system. The high voltage supply was stabilized to about 0.1%. A daily check of the counter sensitivity was made with a standard thallium source. The reactor flux was monitored during irradiation with a neutron counter. A schematic diagram of the equipment is shown in Fig. 2.

Since we are interested only in the variation of the activation ratio with neutron energy, we have not attempted to obtain absolute values; however, the values which are listed in Table I differ only slightly from the absolute activation ratio since only small corrections have been neglected; e.g., the absorption

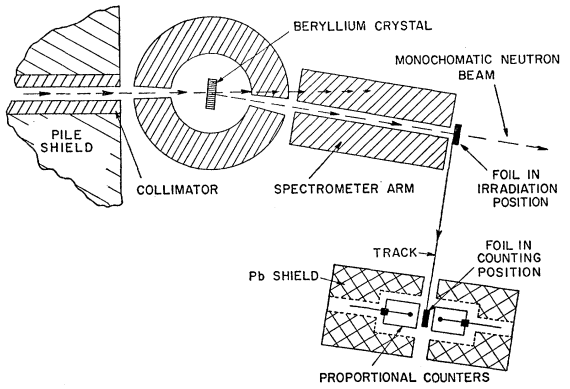


FIG. 2. Schematic diagram of equipment. Monochromatic neutrons are obtained by Bragg reflection from a beryllium crystal. The foil can be shifted from irradiation position to counting position very precisely and rapidly by a slide arrangement.

in the counter windows, the finite sensitivity of the counter to gamma rays, and backscattering.

The correction for self-absorption of the beta particles in the indium foils was important because of the energy difference of the beta spectra. This correction depended on the capture cross section and hence on the neutron energy. The effective capture cross section varied from a maximum of about 30 000 barns at the peak of 1.456-ev resonance to a minimum of about 30 barns at 2.66 ev. For irradiations at an energy where the capture cross section was very large the $In^{116}$ activity was confined to a very thin layer on the front surface of the foil, while at energies where the cross section was small the activity was more uniformly distributed through the foil. A series of measurements were made to investigate the magnitude of the self-absorption effect by using foils of various thickness, and by counting the front-back surfaces of the foils separately. The results are discussed in Sec. IV.

At each neutron energy series of irradiations were made to determine the saturated activities of the

TABLE I. The $In^{116}$ activation ratio. The ratio of 54-minute to 13-second activities, $R(E_i)$, produced by the capture of neutrons in $In^{115}$ are listed for several neutron energies. Resonances occur in the neutron cross section at 1.456 and 3.86 ev. The radiation widths, $\Gamma_\gamma$, of these resonances are listed in Column 2. The total angular momentum, $J$, of the two compound states are listed in Column 3. The foil thicknesses used for each determination are listed in the last column. Presumably these foils were in each case thin enough to eliminate the effects of self-absorption.

| Neutron energy (ev) | $\Gamma_\gamma$ of resonance (ev) | $J$ of resonance | $R(E_i)$ | Foil thickness mg/cm² |
|---|---|---|---|---|
| 1.456 | 0.072[a] | 5[b] | 2.99±0.15 | 26.8 |
| 3.86 | 0.081[a] | 4[b] | 0.85±0.05 | 96 |
| 2.66 | | | 1.57±0.15 | 96 |
| 0.10 | | | 2.5 ±0.3 | 26.8 |
| "Pile beam" | | | 3.02±0.18 | 26.8 |

[a] Taken from reference 4.
[b] Taken from reference 8.

13-second and 54-minute activities separately. The irradiations and counting times each lasted for two or three half-lives. To follow the 13-second activity it was necessary to use two identical scalers which were alternately connected to the output of the amplifier by an automatic timer. Normally, a 5-second count was taken with each scaler alternately. By this method the 13-second decay curve was easily followed.

Background counting rates were regularly measured to determine the background of the proportional counters, the background activity of the foil prior to irradiation, and the spectrometer background arising from neutrons which reached the foil but which were not Bragg scattered from the monochromator crystal. The latter background was obtained by irradiating the foils with the monochromating crystal turned one degree off the Bragg angle. In all cases the backgrounds were negligibly small

### III. TREATMENT OF THE DATA

Let us define the *activation ratio*,[9] $R(E_i)$, as the ratio of the saturated activity of the 54-minute to the 13-second periods obtained at neutron energy $E_i$. Thus,

$$R(E_i) \equiv A_s(54 \text{ min})/A_s(13 \text{ sec}), \tag{1}$$

where $A_s$ is the saturated activity, which can be calculated from the relationship,

$$A_s = A_0(1-e^{-\lambda\tau_i})^{-1}, \tag{2}$$

where $\lambda$ is the decay constant, $\tau_i$ is the irradiation time, and $A_0$ is the activity of sample at the end of the irradiation period.

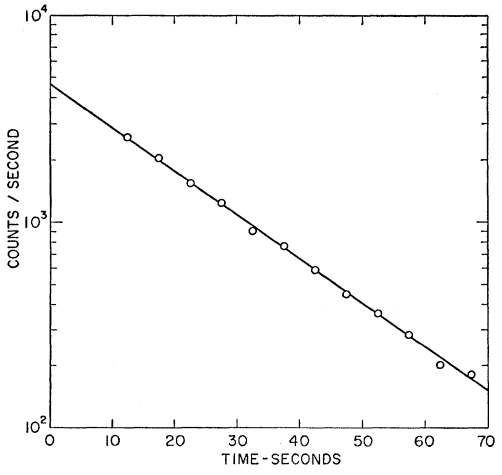


FIG. 3. Decay curve for 13-second activity. Many such sets of data were used for determining the half-life, $T_{\frac{1}{2}}=13.4\pm0.4$ seconds. These data were taken with a 96 mg/cm² foil which was irradiated for 15 seconds in a boron-filtered pile beam.

[9] We shall use the term activation ratio instead of population ratio used by Wood (reference 2), since it seems more appropriate for this case.

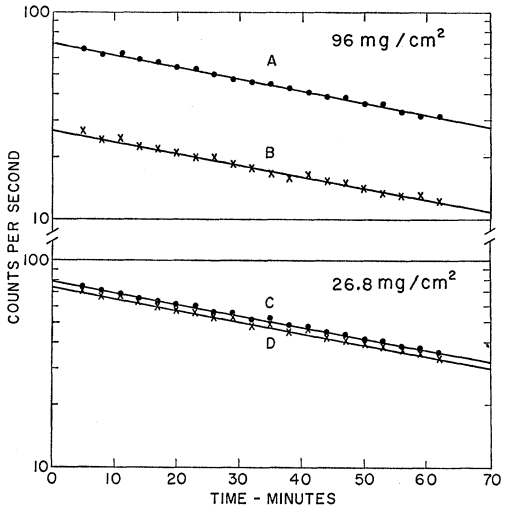


FIG. 4. Effect of foil thickness. Curves A and B on the upper graph show the counting rates obtained at the front-back surfaces respectively of a thick foil. The irradiation was made with 1.456-ev neutrons at which energy the effective cross section is quite large, hence most of the activity is produced toward the front surface of the foil. Self-absorption is large for beta particles emerging from the back surface. Curves C and D are front-back activities obtained with a much thinner foil. Here it is seen that the self-absorption effect is very small. Since the beta particles associated with the 13-second activity are much more energetic than these, the self-absorption is negligible even for the 96 mg/cm² foils.

The counting observations are made at various times after the irradiation has ended; therefore, it is necessary to compute $A_0$ from the observed counts taking proper account of the decay and of the finite counting period. If the counting time is $\Delta t$ and if the counting starts at the time $t_1$ after the end of the irradiation then the number of counts recorded in the counting interval is

$$C\Delta t = K\int_{t_1}^{t_2} -\lambda A_0 e^{-\lambda t}dt, \tag{3}$$

where $C$ is the average counting rate during the interval $\Delta t$, and $t$ is the elapsed time from the end of the irradiation to the middle of the counting interval. The limits of integration are from $t_1=t-\frac{1}{2}\Delta t$ to $t_2=t+\frac{1}{2}\Delta t$. The factor $K$ contains the counter efficiency, geometry, backscattering correction and window absorption. Integration of Eq. (3) gives the result.

$$C = K(2A_0/\Delta t)e^{-\lambda t}\sinh(\tfrac{1}{2}\lambda\Delta t). \tag{4}$$

Combining Eq. (2) and (4) gives

$$A_s = \frac{C\Delta t e^{\lambda t}}{2K(1-e^{-\lambda\tau})\sinh(\tfrac{1}{2}\lambda\Delta t)}. \tag{5}$$

The saturated activities have not been corrected for $K$; however, calculations indicate that to a good approximation the ratio $A_s(54\text{-min})/A_s(13\text{-sec})$ is independent of $K$, except for the effects of self-absorption in the foils which is discussed in the next section.

## IV. MEASUREMENTS AND RESULTS

### A. Decay Constants

Although the decay constants of the $In^{116}$ activity have been measured many times,[10] there is considerable scatter in the reported values. We found it desirable to remeasure the half-lives for use in computing the saturated activities.

For this determination foils of 96 and 26.8 mg/cm² were irradiated in the pile beam and at 0.1 and 0.064 ev where the intensity was great enough to give good counting statistics. Short irradiation times varying from 15 to 45 seconds were used to obtain the 13-second half-life to avoid the build-up of the 54-minute activity. Longer irradiations, 5 to 15 minutes, were used to obtain the 54-minute half-life. A typical decay curve of the 13-second activity is shown in Fig. 3. After minor corrections were made for foil background the counter dead-time, the half-lives were determined from the decay curves by the method of least squares. Many such determinations were averaged to give the values, $T_{\frac{1}{2}}^{*}=53.9\pm0.2$ minutes and $T_{\frac{1}{2}}=13.4\pm0.4$ seconds.

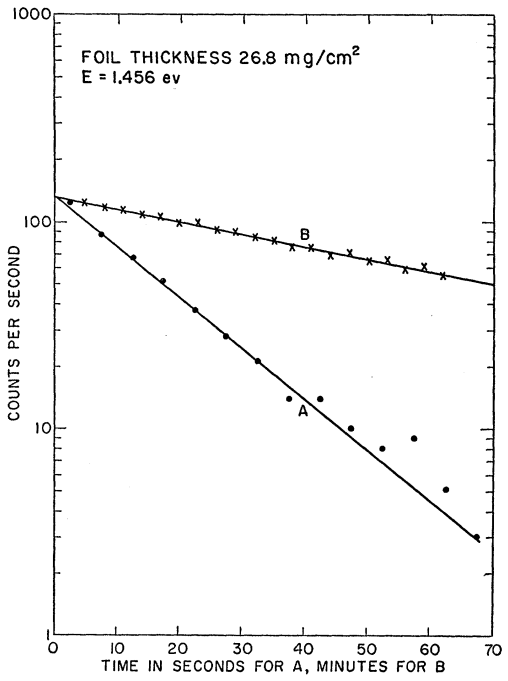


FIG. 5. Typical data set for irradiation energy of 1.456 ev. Curve A shows the 13-second activity in a 26.8 mg/cm² foil resulting from a 42-second irradiation. Curve B is the 54-minute activity obtained in the same foil after a 27-minute irradiation. Saturated activities were computed from these data by the method of least squares. Many such data sets were averaged to obtain the final values.

[10] The data are summarized in the table of D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958), p. 692.

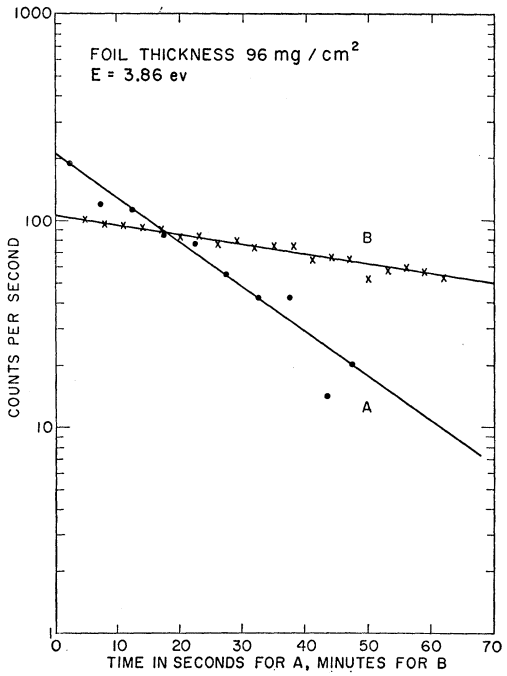


FIG. 6. Typical data set for irradiation energy of 3.68 ev. Curve A shows the 13-second activity in a 96 mg/cm² foil resulting from a 60-second irradiation. Curve B is the 54-minute activity obtained in the same foil after a 54-minute irradiation. Saturated activities were computed from these data by the method of least squares. Many such data sets were averaged to obtain the final values.

### B. Activation Ratios

One of the major difficulties in determining the activation ratio was to select a sample which was thin enough to eliminate the effects of self-absorption. At each energy, a series of foils of various thicknesses were irradiated and the front and back surfaces were counted separately. The final samples which were selected for determining $R(E_i)$ were made thin enough so that the counting rates from the two sides of the foil were practically identical, and so that the same activation ratio was observed for the front and the back (see Fig. 4).

An example of the magnitude of the self-absorption effect, the data obtained at 1.456 ev should be examined. At this energy, a foil thickness of 96 mg/cm² yielded counting rates from the front surface which were about 2.6 times those from the back; however, for a 26 mg/cm² foil the counting rates differed only by about 6%. The 96 mg/cm² foils yielded a value of $R(E_i)$ of 1.985 compared with 2.99 for 26.8 mg/cm² foils. This difference is, of course, due to the selective absorption of the 54-minute $\beta$ particles, since most of the activation occurs near the front surface of the foil because of the very large resonance cross section.

Activations made in the direct pile beam showed a similar spread in values giving $R(E_i)$ of 1.705 and 3.02 for 96 and 26.8 mg/cm² foils, respectively. This spread

is not fully understood, because the thermal cross section which accounts for roughly 85–90% of the activity is not excessively large. Unfortunately this disturbing point was not recognized soon enough to make a proper investigation.

At 2.66 and 3.86 ev the 96 mg/cm² foils proved adequately thin. Measurements indicate that the foil thickness selected for each energy (listed in Table I) approximated "infinitely thin" foils for purposes of obtaining $R(E_i)$

It was necessary to correct the 3.86-ev data for the tail of the huge 1.456-ev resonance. The overlap of the 1.456-ev resonance at 3.86 ev was aggravated by the poor resolution of the crystal spectrometer. The correction was obtained by a series of irradiations at intermediate energies which permitted the tail of the 1.456-ev resonance to be extrapolated. No correction was needed for the overlap of the 3.86-ev resonance at 1.456 ev, since this was completely negligible.

The saturated activities at each neutron energy were computed by the method of least squares from many decay curves using each point on each decay curve. All points were appropriately weighted in this computation to account for counting statistics. Typical sets of data are shown in Figs. 5 and 6.

The activation ratios are summarized in Table I. The cross section for the off-resonance energies (0.100 ev and direct beam) are dominated by the 1.456-ev resonance, and hence yield an activation ratio characteristic of that resonance. The point at 2.66 ev receives contributions from both resonances and therefore has an intermediate value.

The difference in the activation ratio for the two resonances shows that the two compound states decay by quite different gamma-ray cascades. Despite the large number of intermediate excited states which are "accessible" in the cascade process, it appears that a spin difference of one unit between the original two states has a very significant effect on the ultimate populating of the ground and isomeric states. One would expect, therefore that the capture gamma-ray spectra yielded by the two resonances would be markedly different.

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