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**Anomalous  $^{96}\text{Zr}$  resonance integral to thermal activation cross-section ratio and the neutron activation resonance integral of  $^{94}\text{Zr}$  and  $^{96}\text{Zr}$**

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# Anomalous $^{96}\text{Zr}$ resonance integral to thermal activation cross-section ratio and the neutron activation resonance integral of $^{94}\text{Zr}$ and $^{96}\text{Zr}$

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Values of the reduced activity resonance integral relative to the thermal cross section,  $I'/\sigma_0$ , of  $^{96}\text{Zr}$  and  $^{94}\text{Zr}$  were determined relative to gold by measuring cadmium ratios in a reactor spectrum.

Due to the anomalous characteristics of  $^{96}\text{Zr}$  a measurement of  $I'/\sigma_0$  for  $^{96}\text{Zr}$  was undertaken in a "quasi" thermal spectrum.

$^{96}\text{Zr}$  and  $^{94}\text{Zr}$  activation resonance integrals were also determined by measuring the ratio of effective activation cross section of zirconium to  $^{55}\text{Mn}$ . International Atomic Energy Agency (IAEA) calibrated gamma sources were used to determine the samples' absolute activities. A lithium-drifted germanium  $\gamma$ -ray spectrometer was used in all cases to resolve the activities of the irradiated samples.

The results for  $^{96}\text{Zr}$  are  $I'/\sigma_0 = 879 \pm 96$ ,  $I' = 4.97 \pm 0.50$  b, and  $\sigma_0 = 5.7 \pm 1.0$  mb; for  $^{94}\text{Zr}$   $I'/\sigma_0 = 5.87 \pm 0.05$ ,  $I' = 0.369 \pm 0.037$  b, and derived  $\sigma_0 = 63 \pm 8$  mb.

$I'/\sigma_0$  for  $^{96}\text{Zr}$  is an order of magnitude or more greater than that found for stable isotopes. This suggests that almost 100% of the activation resonance integral of  $^{96}\text{Zr}$  consists of  $p$ -neutron captures. This fact is not consistent with the present knowledge of  $p$ -strength functions in the  $90 < A < 130$  mass region.

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## Introduction

The interest in the determination of resonance integrals of zirconium isotopes arises from the importance that the knowledge of zirconium neutron cross sections has in reactor physics. In addition, resonance integrals and  $I'/\sigma_0$  ratios are useful to check the neutron resonance parameters of zirconium, in the mass region  $90 < A < 130$ , where the experimental  $s$ -wave neutron strength functions are anomalously small as far as the original optical model is concerned. Furthermore, the existence in the same mass region of a maximum of the  $p$ -wave neutron strength function predicts an unusually high contribution of  $p$ -neutron captures in the activation resonance integral (Newson 1966).

## Method

The reduced activation resonance integral,  $I'$ , relative to the thermal activation cross section,  $\sigma_0$ , was measured using the cadmium ratio technique.

The  $^{96}\text{Zr}$ -cadmium ratio was measured in a "reactor spectrum" by two techniques. One consisted in measuring the ratio of  $^{96}\text{Zr}$ -cadmium ratio to gold-cadmium ratio, with mixed samples prepared from a very dilute solution containing zirconium and the standard. After irradiation of a bare sample and a cadmium-covered sample, their activities were measured with a lithium-drifted germanium  $\gamma$ -ray spectrometer which makes it possible to measure simultaneously the

standard and investigated photopeaks. This technique has been explained in detail in previous work (Ricabarra *et al.* 1968, 1969).

Due to the special characteristics of  $^{96}\text{Zr}$ -cadmium ratio, a cadmium ratio measurement, using metallic zirconium foils, was also undertaken in the "reactor spectrum" and in a "quasi-thermal spectrum". The difference between these techniques is that in the first one the cadmium ratio is independent of flux gradients, sample weight, and slightly different counting positions, while in the second technique small flux differences between bare and cadmium-covered foils must be carefully corrected.

The  $^{94}\text{Zr}$ -cadmium ratio was measured in the "reactor spectrum" relative to  $^{96}\text{Zr}$ , using metallic zirconium foils.

From the cadmium ratios of zirconium and the epithermal neutron index ( $r(T/T_0)^{1/2}$ ) we obtain  $I'/\sigma_0$  or  $s_0$  in the Westcott formalism (Westcott *et al.* 1958):

$$s_0 = (2/\sqrt{\pi}) I'/\sigma_0$$

The experimental results were corrected by  $G_r$ , the neutron epithermal self-shielding factor;  $F$ , which takes into account the transmission of cadmium for resonance neutrons;  $g$ , the ratio of the activation of the detector in a Maxwellian flux to that of a  $1/r$  detector having the same  $\sigma_0$ ;  $W$ , which is the difference of epithermal neutron activation and that of a  $1/r$  detector normalized to  $\sigma_0 = 1$  below the cadmium cutoff; and a spectrum correction factor that takes into account

the deviation from the  $1/E$  neutron spectrum, calculated with multigroup diffusion theory (Boix and Solanilla 1967).

The procedure followed in our previous work was to obtain the value of  $I'$  by normalizing the measured  $I'/\sigma_0$  to the thermal activation cross section quoted by other authors. This procedure was not followed here and a determination of  $I'$  was undertaken because previous measurements

of activation thermal cross section for  $^{94}\text{Zr}$  and  $^{96}\text{Zr}$  have been made in a reactor spectrum (Lyon 1960). The thermal cross section of  $^{55}\text{Mn}$  was used as a standard by measuring the activity of  $^{56}\text{Mn}$  produced by neutron capture.

The ratio of activation per atom of  $^{94}\text{Zr}$  or  $^{96}\text{Zr}$  to  $^{55}\text{Mn}$  can be expressed, in the Westcott formalism, as the ratio of effective cross sections,  $\hat{\sigma}$ .

$$\hat{\sigma}(\text{Zr})/\hat{\sigma}(\text{Mn}) = (2/\sqrt{\pi})(r(T/T_0)^{1/2})(I'G_r/g)_{\text{Zr}} \frac{(1 + ((r(T/T_0)^{1/2})s_0G_r/g)^{-1})_{\text{Zr}}}{(\sigma_0G_{\text{th}})_{\text{Mn}} \left(1 + (r(T/T_0)^{1/2}) \frac{s_0G_r}{gG_{\text{th}}}\right)_{\text{Mn}}}$$

where

$$(r(T/T_0)^{1/2}) \frac{s_0G_r}{gG_{\text{th}}} \equiv \frac{1 - (r(T/T_0)^{1/2})(R_{\text{Cd}}/G_{\text{th}})(1/K - FW/g)}{(FR_{\text{Cd}} - 1)}$$

can be obtained from the measured cadmium ratios,  $R_{\text{Cd}}$ ;  $1/K = 4(E_0/(\pi E_{\text{Cd}}))^{1/2}$ , depends on the cadmium cutoff energy,  $E_{\text{Cd}}$ , and  $E_0 = 0.025$  eV;  $G_{\text{th}}$  takes into account the thermal neutron self-shielding in the foil (Hanna 1963). For Zr and Mn,  $F = 1$ ,  $W = 0$ , and  $g = 1$  were assumed. Rearranging gives

$$[1] \quad I'(\text{Zr}) = \frac{\hat{\sigma}(\text{Zr})/\hat{\sigma}(\text{Mn})}{G_r(\text{Zr})} \frac{(\sigma_0G_{\text{th}})_{\text{Mn}}}{(r(T/T_0)^{1/2})(2/\sqrt{\pi})} \frac{(1 + (r(T/T_0)^{1/2})s_0G_r/G_{\text{th}})_{\text{Mn}}}{(1 + ((r(T/T_0)^{1/2})s_0G_r)^{-1})_{\text{Zr}}}$$

Zirconium  $G_r$  and manganese  $G_{\text{th}}$  are close to unity and have been calculated. For the thin zirconium foils we use  $G_{\text{th}} = 1$ .

The activation ratio per atom is obtained from the specific activities at the end of irradiation,  $Q_0$ , of Zr and Mn:

$$[2] \quad \hat{\sigma}(\text{Zr})/\hat{\sigma}(\text{Mn}) = \frac{Q_0(\text{Zr})}{Q_0(\text{Mn})} \frac{((1 - e^{-\lambda T})p/A)_{\text{Mn}}}{((1 - e^{-\lambda T})p/A)_{\text{Zr}}}$$

where  $\lambda$  is the decay constant,  $T$  the irradiation time,  $A$  the atomic weight, and  $p$  the isotopic concentration or the atom percent of the element when an alloy is used.

#### Determination of $^{96}\text{Zr}$ $I'/\sigma_0$ Ratio

Cadmium ratio irradiations were made in the internal graphite reflector of the "Reactor Argentino 1" (RA1), 2 cm away from the core, where the gold-cadmium ratio is  $R_{\text{Cd}}(\text{Au}) = 1.684 \pm 0.0003$  and the epithermal neutron index  $(r(T/T_0)^{1/2}) = 0.0794 \pm 0.0003$  using the gold parameters given in Table I.

Two samples with  $24 \text{ mg/cm}^2$  zirconium and  $0.02 \text{ mg/cm}^2$  gold were irradiated bare and cadmium covered for 6 h and measured 1 day later.

$^{96}\text{Zr}$  produces by neutron capture  $^{97}\text{Zr}$  (17.0 h) which decays to  $^{97}\text{Nb}^{\text{m}}$  (1 min) and  $^{97}\text{Nb}$  (72.1 min) (Fig. 1). The  $^{97}\text{Nb}^{\text{m}}$  743.3 keV

and  $^{97}\text{Nb}$  658.1 keV gamma rays were measured relative to the  $^{198}\text{Au}$  412 keV gamma ray.

Bare and cadmium-covered samples were counted alternately and the decay was followed for at least 70 h. The half-life of  $^{97}\text{Zr}$  agreed with the value currently quoted in the literature.

The average from two irradiations gives

$$R_{\text{Cd}}(^{96}\text{Zr})/R_{\text{Cd}}(^{197}\text{Au}) = 0.5989 \pm 0.0005$$

or with

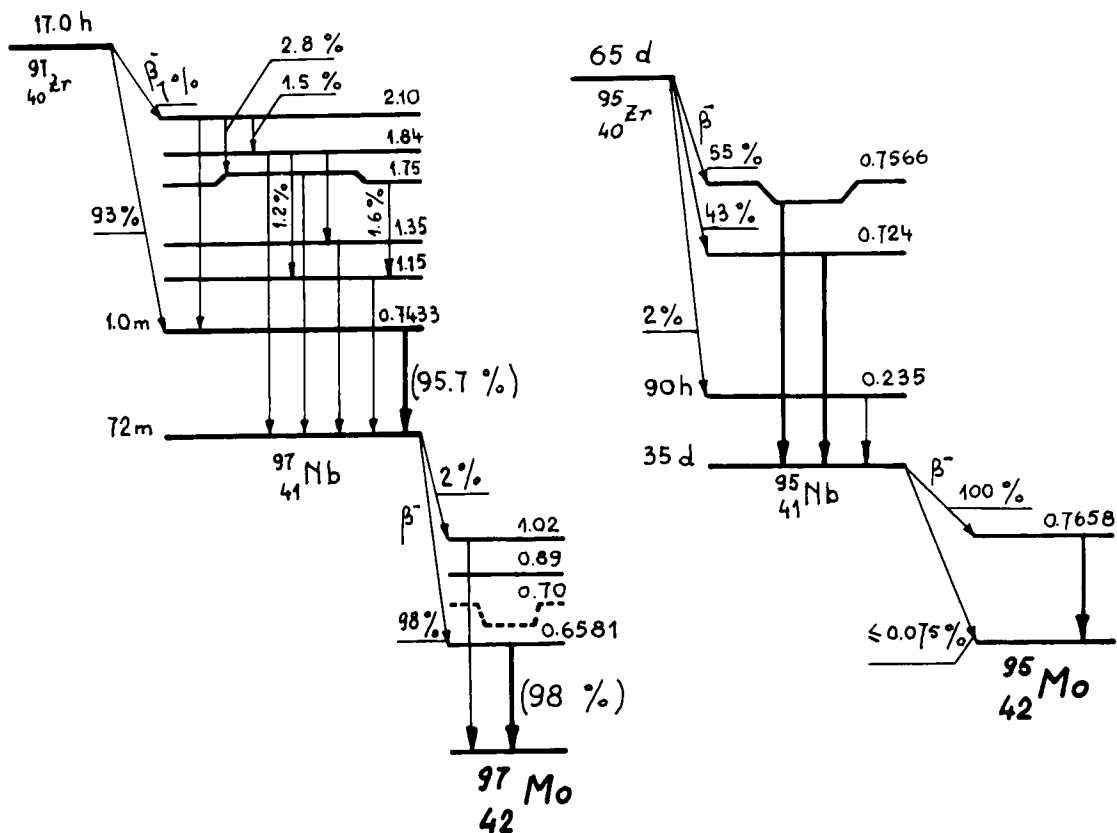
$$R_{\text{Cd}}(^{197}\text{Au}) = 1.684 \pm 0.003$$

$$R_{\text{Cd}}(^{96}\text{Zr}) = 1.0086 \pm 0.002$$

With  $G_r = 0.98$  and a  $-10\%$  spectrum correction, we get

$$S_0 = 1308 \pm 305$$

$$I'/\sigma_0 = 1160 \pm 270$$

FIG. 1. Decay schemes of  $^{97}\text{Zr}$  and  $^{95}\text{Zr}$ .TABLE I  
 $^{115}\text{In}$  and  $^{197}\text{Au}$  parameters

|       | In     | Au     |
|-------|--------|--------|
| $S_0$ | 18.7*  | 17.7   |
| $g$   | 1.028  | 1.0078 |
| $W$   | 0.335  | 0.089  |
| $F$   | 0.960† | 0.998  |
| $1/K$ |        | 0.462  |

\*Experimental value using Au as standard.

†Experimental value determined by measuring indium-cadmium ratios with several cadmium thicknesses.

For a cadmium ratio so near to unity any systematic or statistical error is quite significant in  $R_{\text{Cd}} - 1$ , which is the quantity of interest. To avoid possible errors that may arise from variations in the relative gold to zirconium concentration, which are estimated to be around 0.2%, a direct cadmium ratio measurement was undertaken.

Very thin metallic zirconium foils, 13 mg/cm<sup>2</sup> thick and 99.99% pure, were irradiated in the

“reactor spectrum” position. A cadmium-covered foil was located along the longitudinal axis of the irradiation capsule between two bare foils. The foils’ position was parallel to the longitudinal axis and the distance between foils was 3 cm (the monitor foils used to determine the epithermal component of the flux were irradiated in the same position). The bare activity in the cadmium position was obtained by linear interpolation from the specific activities of the bare foils, and their ratio to the cadmium covered one was taken to obtain the cadmium ratio. The foils were weighed to an accuracy of 0.03%.

As has been described in previous work (Ricabarra *et al.* 1968) the place of irradiation in the “reactor spectrum” position has an ample region with flat epithermal index and epithermal flux and since the samples are irradiated in the middle plane of the reactor, local thermal flux gradients are quite small. This is reflected in the good statistical reproducibility as shown in

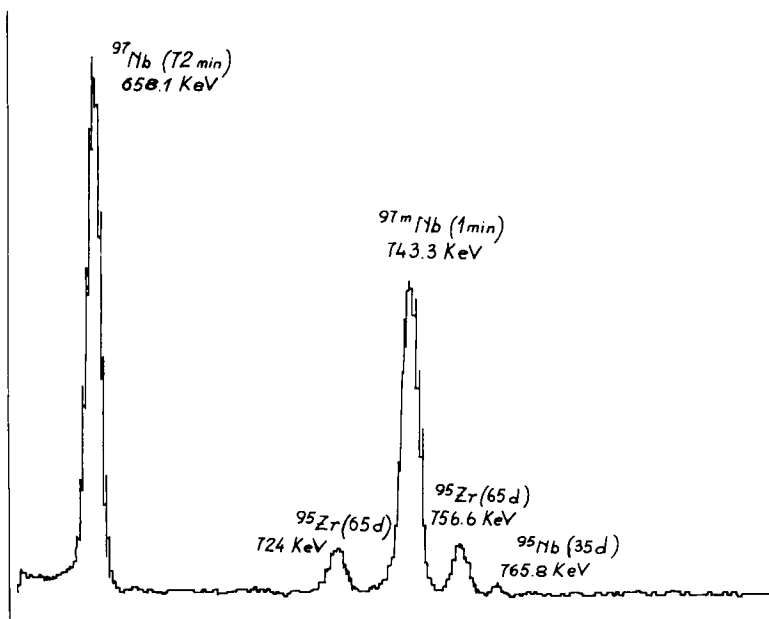


FIG. 2. Gamma-ray spectrum of a zirconium foil, 3 days after irradiation.

TABLE II

 Summary of  $^{96}\text{Zr}$  cadmium ratio measurements in the reactor spectrum with zirconium foils

| Run             | 1                   | 2     | 3     | 4     |
|-----------------|---------------------|-------|-------|-------|
| $R_{\text{Cd}}$ | 1.011               | 1.012 | 1.013 | 1.011 |
| Average         | $1.0118 \pm 0.0005$ |       |       |       |

Table II. The difference in the specific activity between bare foils was around 1% and the error in the linear interpolation can be estimated to be less than 0.1%.

The activity of the 743.3 keV and 658.1 keV  $\gamma$  rays of the zirconium foils was measured with the lithium-drifted germanium  $\gamma$ -ray spectrometer. A typical  $\gamma$ -ray spectrum is shown in Fig. 2. Dead time in the counting system was kept below 0.5% and the multichannel analyzer operated in live-time mode. Since the foils were approximately of the same activity, dead-time corrections were negligible. The results from a number of pairs of spectra with a total of about  $10^6$  counts were used in each ratio measured.

In Table II are listed the results of four independent irradiations. The averaged result gives

$$R_{\text{Cd}}(^{96}\text{Zr}) = 1.0118 \pm 0.0005$$

Including a  $-10\%$  spectrum correction and

resonance self-shielding factor  $G_r = 0.99$ , we get

$$S_0 = 945 \pm 40$$

$$I'/\sigma_0 = 838 \pm 34$$

The quoted error is the standard deviation which is not only an indication of counting statistics but also of small differences that may arise from different irradiation and counting positions.

The contribution of thermal capture is so small that in order to have a confirmation of the  $I'/\sigma_0$  value, we made a determination in a spectrum with 30 times higher thermal to epithermal ratio.

The measurements were made at 21 cm from the core vessel in the graphite reflector which is enlarged to form a thermal column. Diffusion multi-group calculations predict that at about 21 cm from the core we will have a nearly  $1/E$  epithermal neutron spectrum. Figure 3 shows the slowing down neutron spectrum at 2 cm and 21 cm in the graphite reflector calculated with a 54 group diffusion code (Boix and Solanilla 1967).

An indium-cadmium ratio measurement at the irradiation site  $R_{\text{Cd}}(^{115}\text{In}) = 20.3 \pm 0.3$  gives an epithermal neutron index  $(r(T/T_0)^{1/2}) = 0.00294 \pm 0.00005$  (indium parameters in Table I).

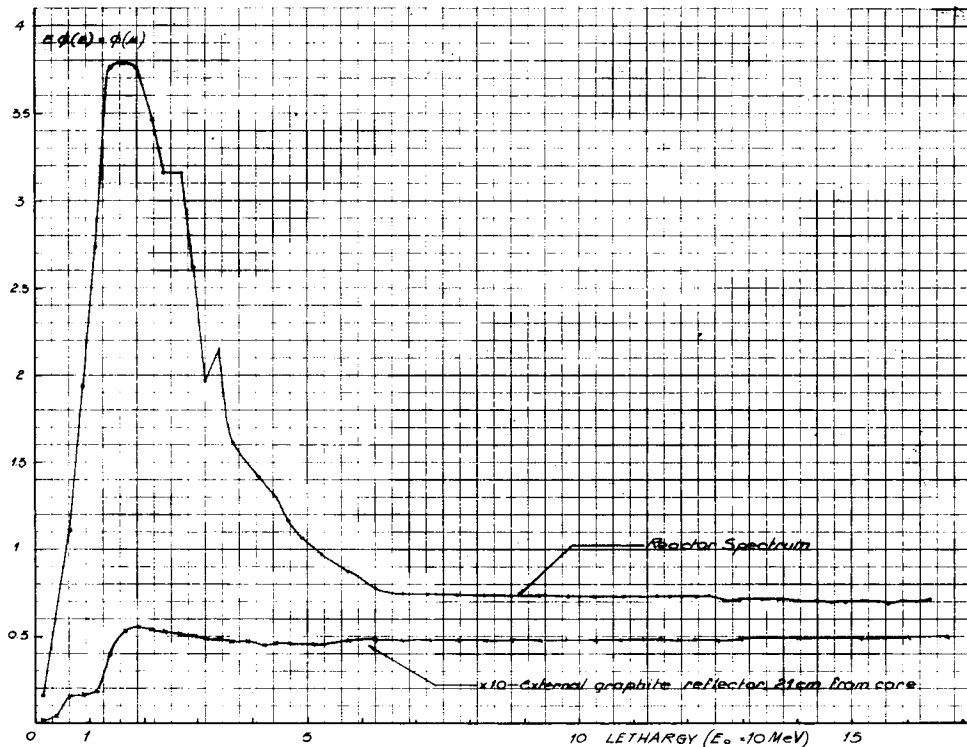


FIG. 3. Spectrum form function  $\phi(u)$ .

The averaged result from four irradiations was

$$R_{cd}(^{96}\text{Zr}) = 1.34 \pm 0.04$$

$$S_0 = 1009 \pm 120$$

$$I'/\sigma_0 = 895 \pm 107$$

The standard deviation is larger than in the "reactor spectrum" measurements, reflecting the fact that reproducibility of foil position in the neutron field was not as good, because the gradients of neutron flux in the "21 cm" position were quite large.

In Table III are summarized the results from the three techniques; the agreement is good within the errors.

The weighted average is

$$S_0 = 992 \pm 108$$

$$I'/\sigma_0 = 879 \pm 96$$

#### Determination of $^{96}\text{Zr}$ Activation Resonance Integral

A determination of the activation resonance integral of  $^{96}\text{Zr}$  was undertaken using the  $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$  reaction as the standard.

A thin zirconium metallic foil and a 96.3 mg/cm<sup>2</sup> manganese foil (Johnson and Matthey, 88% Mn and 12% Ni) were irradiated in the internal graphite reflector in exactly the same position where  $I'/\sigma_0$  was measured. Approximately 12 h after irradiation the 658.1 keV  $^{97}\text{Nb}$  (72 m)  $\gamma$  ray was measured relative to the 661.6 keV  $^{137}\text{Cs}$   $\gamma$  ray of an IAEA calibrated source (standard error 2%), with a lithium-drifted germanium  $\gamma$ -ray spectrometer.

$^{97}\text{Zr}$  decays to  $^{97}\text{Nb}$ ; 98% of  $^{97}\text{Nb}$   $\beta$  decay feeds the 658.1 keV level; 93.5% of  $^{137}\text{Cs}$  decays to the 661.6 keV level of  $^{137}\text{Ba}^m$  ( $e/\gamma = 0.110$ ).

Then it is easy to obtain the activity of  $^{97}\text{Zr}$  from  $^{97}\text{Nb}$ , correcting for the small differences in counting efficiency (0.87%) between the  $^{97}\text{Nb}$  658.1 keV and the  $^{137}\text{Cs}$  661.6 keV  $\gamma$  rays and a factor (1.077) that allows for the combined decay of  $^{97}\text{Zr}$  (17.0 h) and  $^{97}\text{Nb}$  (72 min).

The absolute activity of the manganese foil was measured by  $4\pi\beta$  counting or by gamma activity intercalibration.

Beta counting of  $^{56}\text{Mn}$  ( $2.756 \pm 0.002$  h) (Bartholomew *et al.* 1953) was made in a  $4\pi$  methane flow proportional counter (400 V

TABLE III  
Comparison of  $^{96}\text{Zr}$   $I'/\sigma_0$  measurements

| Irradiation site                            | Thickness (mg/cm <sup>2</sup> ) | $r(T/T_0)^{1/2}$      | $G_r$ | $I'/\sigma_0$  | $S_0$          |
|---|---------------------------------|-----------------------|-------|----------------|----------------|
| Reactor spectrum<br>(2 cm from core)        |                                 |                       |       |                |                |
| (a) Mixed samples                           | 24                              | $0.0794 \pm 0.0003$   | 0.98  | $1160 \pm 270$ | $1308 \pm 305$ |
| (b) 0.02 mm thick zirconium foils           | 13                              | $0.0794 \pm 0.0003$   | 0.99  | $838 \pm 34$   | $945 \pm 40$   |
| Quasi thermal spectrum<br>(21 cm from core) | 13                              | $0.00294 \pm 0.00005$ | 0.99  | $895 \pm 107$  | $1009 \pm 120$ |
| Average                                     |                                 |                       |       | $879 \pm 96$   | $992 \pm 108$  |

TABLE IV  
Summary of  $^{96}\text{Zr}$  reduced neutron activation resonance integral measurements

|  | Thickness (mg/cm <sup>2</sup> ) | $G_r$ | $\hat{\sigma}(^{96}\text{Zr})/\hat{\sigma}(^{55}\text{Mn})$ | $I'$ (b)                 |
|--|---------------------------------|-------|---|--------------------------|
| $^{96}\text{Zr}$ gamma counting and $^{56}\text{Mn}$ beta counting | 13.0                            | 0.99  | 0.03548<br>0.03545<br>0.03832<br>0.03451                    |                          |
| Average  |                                 |       | $0.03594 \pm 0.00082^*$                                     | $5.102 \pm 0.45^\dagger$ |
| $^{96}\text{Zr}$ and $^{56}\text{Mn}$ gamma counting               | 39.0                            | 0.97  | 0.03412<br>0.03272  |                          |
| Average  |                                 |       | $0.03342 \pm 0.00070^*$                                     | $4.842 \pm 0.49^\dagger$ |

\*The quoted error is the standard deviation.

†The quoted error takes into account the standard deviation from the experiment plus the estimated systematic errors discussed in the text.

plateau, 0.1% slope). Beta self-absorption is the most important factor we must take into account in order to have the true foil activity. A careful measurement of the effective self-absorption for Mn-Ni foils has been made in a  $4\pi$  counter operated in the Geiger region by Meister (1958), for the foil thickness used by us  $96.3 \text{ mg/cm}^2$ ,  $S_\beta = 0.721 \pm 0.014$ . Small correction factors due to beta absorption in the mylar support of the  $4\pi$  counter, dead-time correction, etc. have been taken into account (Patte and Yaffe 1955, 1956; Mann and Seliger 1953).

In two experiments out of six the  $^{56}\text{Mn}$  846.9 keV  $\gamma$  ray (98.9%) was measured relative to the 834.9 keV  $\gamma$  ray of the  $^{54}\text{Mn}$  IAEA calibrated source. The results were corrected by the difference in efficiency of the  $^{56}\text{Mn}$  and  $^{54}\text{Mn}$   $\gamma$  rays in the lithium-drifted germanium detector (3.1%).

The  $^{137}\text{Cs}$ (30 years(y)) and  $^{54}\text{Mn}$ (303 day(d)) IAEA calibrated sources were corrected for decay from calibration date (1-1-1968) to that of the measurement.

$I'$  was obtained from  $\hat{\sigma}(^{96}\text{Zr})/\hat{\sigma}(^{56}\text{Mn})$  (expression [1]) using  $\sigma_0(^{55}\text{Mn}) = 13.3 \pm 0.1 \text{ b}$  (Goldberg *et al.* 1966),  $G_{\text{th}} = 0.997$ ,  $R_{\text{cd}}(^{55}\text{Mn}) = 11.2 \pm 0.1$ , and a  $-10\%$  spectrum correction.

The results from six experiments are listed in Table IV.

The error quoted for the resonance integral is the standard deviation from the measurements plus the estimated systematic errors.

In the measurement of  $^{96}\text{Zr}$  the systematic errors which were taken into account are: 2% quoted error for calibration of the IAEA source and 1% due to differences in counting position of the IAEA source and the zirconium foil. The error in the  $4\pi$  counting of  $^{56}\text{Mn}$  is due mainly to the error in the beta self-absorption correction which was estimated to be 2%. Other errors are not considered because they are much lower than the uncertainty in the beta self-absorption factor.

The  $^{56}\text{Mn}$  comparison with the  $^{54}\text{Mn}$  IAEA source has a 1% quoted error in the standard calibration, 1% from the difference between the foil and the IAEA standard source position, and

TABLE V  
 $^{96}\text{Zr}$  thermal cross section calculated from resonance parameters

| $E$<br>(eV)                             | $\Gamma_n$<br>(eV) | $\Gamma_\gamma$<br>(eV) | $\sigma_0$<br>(mb) | References          |
|---|--------------------|-------------------------|--------------------|---------------------|
| 300.9                                   | 0.231              | 0.220                   | 133                | Morgenstern (1968)  |
| 2680                                    | 4                  | 0.300*                  | 13.2               | Good and Kim (1968) |
| 3850                                    | 7                  | "                       | 9.36               |                     |
| 4110                                    | 13                 | "                       | 14.8               |                     |
| 4610                                    | 6                  | "                       | 5.11               |                     |
| 5480                                    | 15.6               | "                       | 8.63               |                     |
| 5840                                    | 4                  | "                       | 1.89               |                     |
| 5970                                    | 4                  | "                       | 1.79               |                     |
| 6800                                    | 4                  | "                       | 1.29               |                     |
| The sum of higher energy resonances is  |                    |                         | 10                 |                     |
| The calculated thermal cross section is |                    |                         | 199 mb             |                     |

\*Bartolome *et al.* (1969).

3% error in extrapolating the decay of the standard source for about two half-lives due to uncertainties in the  $^{54}\text{Mn}$  half-life quoted in the literature.

Common to both methods of calibration is a 0.5% assumed error in the chemical composition of the manganese-nickel alloy, a 0.7% error introduced by the error in the  $^{56}\text{Mn}$  half-life by extrapolating about 30 h to the end of the irradiation and 0.8% in the  $^{56}\text{Mn}$  thermal cross section. The average value from both methods is

$$I' = 4.97 \pm 0.50 \text{ b}$$

where the quoted error takes into account 2% from the standard deviation plus our best estimation of the systematic errors. From our measured  $I'/\sigma_0$  and  $I'$ , the activation thermal cross section for  $^{96}\text{Zr}$ ,  $\sigma_0 = 5.7 \pm 1.0 \text{ mb}$ , can be derived.

Calculation of  $I'$  from the neutron resonance parameters of  $^{96}\text{Zr}$  was made using  $\Gamma_n = 0.231 \text{ eV}$  and  $\Gamma_\gamma = 0.220 \text{ eV}$  (Morgenstern 1968) for the 300.9 eV resonance and for resonances above 2 keV, the neutron resonance widths from Good and Kim (1968), and an average radiation width,  $\Gamma_\gamma = 0.300 \text{ eV}$ , measured by Bartolome *et al.* 1969 for the other zirconium isotopes. The calculated  $I' = 5.61 \pm 0.90 \text{ b}$  is in reasonable agreement with our measured value. On the other hand, calculated  $I'/\sigma_0$  and  $\sigma_0$  values are in disagreement with the experimental values by almost two orders of magnitude (Table V).

#### $I'/\sigma_0$ and Reduced Activation Resonance Integral Determination of $^{94}\text{Zr}$

The cadmium ratio of  $^{94}\text{Zr}$  was measured relative to  $^{96}\text{Zr}$ , waiting approximately 6 d after

irradiation in order to have the same order of activity in the  $\gamma$  rays of both isotopes.

$^{94}\text{Zr}$  by neutron capture produces  $^{95}\text{Zr}$  (65 d) which by beta decay feeds the 756.6 keV and 724 keV levels; these correspond to 55% and 43% (Lederer *et al.* 1968), respectively, of the total disintegration rate. The 756.6 keV and the 724 keV  $\gamma$  rays were measured relative to the 658.1 keV and 743.3 keV  $\gamma$  rays from decay of  $^{96}\text{Zr}$  (Fig. 1).

The average result from five irradiations is

$$R_{\text{Cd}}(^{94}\text{Zr})/R_{\text{Cd}}(^{96}\text{Zr}) = 2.498 \pm 0.012$$

and with

$$R_{\text{Cd}}(^{96}\text{Zr}) = 1.0118$$

$$R_{\text{Cd}}(^{94}\text{Zr}) = 2.527 \pm 0.013$$

Including a -13% spectrum correction and with epithermal corrections  $G_r = 1$  and  $F = 1$ , we get

$$S_0(^{94}\text{Zr}) = 6.62 \pm 0.06$$

$$I'/\sigma_0(^{94}\text{Zr}) = 5.87 \pm 0.05$$

A  $^{94}\text{Zr}$  cross-section measurement by Lyon (1960) was made in a reactor spectrum. This effective cross section  $\hat{\sigma} = 75 \text{ mb}$  can be corrected, using an epithermal neutron index deduced from the  $^{55}\text{Mn}$ - and  $^{60}\text{Co}$ -cadmium ratio quoted by Lyon and our measured  $I'/\sigma_0$  value, to obtain  $\sigma_0 = 58 \text{ mb}$ . From this  $I' = 0.350 \text{ b}$  is obtained.

In order to confirm this value we undertook an absolute determination of  $I'$  relative to  $^{55}\text{Mn}$  using  $^{96}\text{Zr}$  as a secondary standard. The sum of 724 keV and 756.6 keV  $\gamma$ -ray activities (98% of

$^{95}\text{Zr}$  beta decay) was measured relative to the 743.3 keV  $^{97}\text{Nb}^m$  photopeak which represents 95.7% of the total  $^{96}\text{Zr}$  disintegrations (Fig. 1).

The average from two runs gives

$$\hat{\sigma}(^{94}\text{Zr})/\hat{\sigma}(^{96}\text{Zr}) = 0.199 \pm 0.006$$

and with

$$\hat{\sigma}(^{96}\text{Zr})/\hat{\sigma}(^{55}\text{Mn}) = 0.0359 \pm 0.0008$$

$$\hat{\sigma}(^{94}\text{Zr})/\hat{\sigma}(^{55}\text{Mn}) = 0.0714 \pm 0.0027$$

and

$$I'(^{94}\text{Zr}) = 0.369 \pm 0.037$$

The errors in  $I'$  take into account the experimental standard deviation plus 8% of the estimated systematic errors. Discussion of systematic errors is similar to the  $^{96}\text{Zr}$  and will not be repeated here.

A thermal cross section,  $\sigma_0 = 0.063 \pm 0.008$  b, can be derived from our experimental values.

The  $^{94}\text{Zr}$  resonance integral was calculated using neutron resonance parameters measured by Bartolome *et al.* (1969) between 2.2 keV and 14.2 keV; for higher energies the values from Good and Kim (1968) were used.

The calculated value is  $I' = 0.284$  b, in reasonable agreement with the experimental value. The calculated thermal cross section (excluding  $p$ -wave neutron resonances) gives 44.3 mb. Comparing the calculated  $I'/\sigma_0 = 6.41$  with the experimental value, the agreement is much better.

Calculation of the resonance integral shows that about half of the activation comes from captures in the  $p$ -wave neutron resonances in the keV energy region of  $^{94}\text{Zr}$ .

### Discussion and Conclusion

Our qualitative studies of cadmium ratios of about 50 isotopes and quantitative determination of 25 isotopes and the critical review of previous results in other isotopes (Persiani 1963; Goldberg *et al.* 1966) show that  $I'/\sigma_0$  values are usually lower than 100.

The highest  $I'/\sigma_0$  values found in our literature search correspond to  $^{124}\text{Sn}$  and  $^{128}\text{Xe}$  (Tilbury and Kramer 1968) in which, according to the approximate cadmium ratios quoted by the authors,  $I'/\sigma_0$  values may be around 100.  $^{124}\text{Sn}$  is characterized by having a  $p$ -wave neutron resonance at 62 eV which is around 100 times

stronger than expected for an average  $p$ -wave resonance for this nuclide (Harvey and Fuketa 1965).

Then we may say that the experimental value of  $I'/\sigma_0$  for  $^{96}\text{Zr}$  is by an order of magnitude the highest that has been found in stable isotopes.

The value of  $I'/\sigma_0$  calculated with the neutron resonance parameters of  $^{96}\text{Zr}$  (Good and Kim 1968; Morgenstern 1968) gives a value almost an order of two lower and even assuming that the 301 eV resonance is a  $p$ -wave neutron resonance, calculated  $I'/\sigma_0$  gives a value an order of one lower than the experimental one.

In Table V the calculated contribution to the thermal absorption cross section by every resonance of  $^{96}\text{Zr}$  is listed. It can be observed that only the contribution of one of the keV energy  $s$ -wave resonances is enough to explain the magnitude of the experimental thermal cross section.

On the other hand, the excellent agreement between the experimental and calculated resonance integral shows that there is no serious error in the  $^{96}\text{Zr}$  neutron resonance parameters.

Then the present measurement would suggest that first the 301 eV resonance of  $^{96}\text{Zr}$  is a  $p$ -wave resonance and second that some of the resonances in the keV energy region which were analyzed by Good and Kim as  $s$ -wave neutron resonances are probably  $p$ -wave resonances.

This suggestion cannot be reconciled with the analysis of average neutron resonance parameters of  $^{96}\text{Zr}$  made by Good and Kim (1968) or with the values of the  $p$ -strength function from nearby even-even zirconium isotopes (Bartolome *et al.* 1969) because a  $p$ -strength function for  $^{96}\text{Zr}$  would have to be considered around  $30 \times 10^{-4}$  if our suggestions were correct.

Furthermore, the excellent agreement observed between our experimental and calculated results of  $I'/\sigma_0$  and  $I'$  for  $^{94}\text{Zr}$  enhances still more the difficulty of explaining the anomalous behavior of the  $^{96}\text{Zr}$   $I'/\sigma_0$  ratio.

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