

The resonance integrals of  $^{63}\text{Cu}$ ,  $^{65}\text{Cu}$ ,  $^{107}\text{Ag}$ ,  $^{159}\text{Tb}$ ,  $^{164}\text{Dy}$  and  $^{165}\text{Ho}$

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# The resonance integrals of $^{63}\text{Cu}$ , $^{65}\text{Cu}$ , $^{107}\text{Ag}$ , $^{159}\text{Tb}$ , $^{164}\text{Dy}$ and $^{165}\text{Ho}$

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**Abstract.** The reduced resonance integrals were measured by an activation technique using cadmium ratios, relative to two standard materials, manganese and gold. The use of two reference standards corrects for small deviations in the neutron slowing down spectrum from a  $1/E$  dependence per unit neutron energy  $E$ . Results obtained were  $^{63}\text{Cu}$ :  $2.79 \pm 0.18$  b,  $^{65}\text{Cu}$ :  $1.22 \pm 0.06$  b,  $^{107}\text{Ag}$ :  $79 \pm 4$  b,  $^{159}\text{Tb}$ :  $390 \pm 24$  b,  $^{164}\text{Dy}$ :  $-514 \pm 115$  b and  $^{165}\text{Ho}$ :  $618 \pm 33$  b, where the uncertainties are estimated at approximately the 99% confidence level, and include an uncertainty of  $\pm 4\%$  in the reference standards.

It was necessary to measure the half-lives of the activities produced, giving  $^{64}\text{Cu}$ :  $12.704 \pm 0.006$  h,  $^{66}\text{Cu}$ :  $5.13 \pm 0.03$  min,  $^{108}\text{Ag}$ :  $2.37 \pm 0.01$  min and  $^{168}\text{Ho}$ :  $26.83 \pm 0.03$  h, and also the  $2200 \text{ ms}^{-1}$  neutron capture cross sections of  $^{159}\text{Tb}$ :  $23.2 \pm 0.5$  b,  $^{165}\text{Ho}$ :  $61.2 \pm 1.1$  b and  $^{107}\text{Ag}$ :  $37.6 \pm 1.2$  b (re-evaluated from the new half-life).

## 1. Introduction

Resonance integrals are required in experimental reactor physics, in the measurement of neutron slowing down spectra, in some dosimetry applications, in neutron standardization measurements (eg in flux and neutron source strength measurements) and in checking nuclear resonance parameters. For most isotopes a wide range of reported values exists, so that often a resonance integral is not known within 20%, making it of little practical use. It is also apparent that the uncertainties quoted for many measurements are poorly defined and unrealistically small. As the required precision of measurements increases, it becomes desirable to provide more consistent resonance integral data.

Methods of measurement are nearly always by the pile reactivity or activation techniques, and results from these two methods should be compatible, when appropriate corrections for neutron spectra, resonance self-shielding and scattering and competing nuclear reactions are made, but often the discrepancies are unacceptably large. We have used a simple activation technique employing cadmium ratio measurements, where resonance integrals are measured in a slowing down flux which is closely proportional to  $1/E$  ( $E$  is neutron energy) relative to two selected standard materials, Mn and Au, whose resonance integrals are assumed known to  $\pm 4\%$ . The use of two standard materials with well separated principal resonance energies (4.9 and 337 eV) partially corrects for small deviations in the epithermal neutron spectrum from a  $1/E$  dependence, especially for isotopes whose principal resonances lie within this energy interval. Measurements yield values of  $I'/\sigma_0$ , where  $I'$  is the reduced resonance integral and  $\sigma_0$  is the  $2200 \text{ ms}^{-1}$  neutron capture cross section. If  $\sigma_0$  is known to about 3%, we can expect an accuracy in  $I'$  of about 6%. During the course of the work it was necessary to measure

the half-lives of some of the activities (for the analysis of the decay data) and the neutron capture cross sections of Ho and Tb. The reduced resonance integrals are compared with literature values, but only in the case of the Cu isotopes have these been measured previously with comparable accuracy.

## 2. Theory

The reduced resonance integral is

$$I' = \int_{E_c}^{\infty} \left[ \sigma_a(E) - g\sigma_0 \left( \frac{E_0}{E} \right)^{1/2} \right] \frac{dE}{E} \quad (1)$$

where  $\sigma_a(E)$  is the neutron capture cross section at neutron energy  $E$ , and suffix 0 refers to a neutron velocity of  $2200 \text{ ms}^{-1}$ ,  $g$  is the Westcott *et al* (1958) factor which accounts for the deviation of the cross section from a  $E^{-1/2}$  behaviour in the thermal region.  $E_c$  is the effective epithermal cut-off energy, which depends on the temperature  $T$  of the Maxwellian thermal neutron flux distribution and on the moderator. The value of  $I'$  is not strongly dependent on the exact value chosen for  $E_c$  (about 0.1 eV) for the isotopes considered here because their principal resonances are at much higher energies, except for  $^{164}\text{Dy}$ .

It is assumed that in an aqueous moderator the epithermal neutron flux is closely proportional to  $1/E$  per unit energy in the resonance region, and that this shape is not influenced by the foil holder. Thus the usual Westcott *et al* flux convention can be applied without any corrections for the deviation of the spectrum shape. The bare foil activity per atom per unit flux density may be written as

$$A = \sigma_0(gG_t + G_r x I' / \sigma_0) \quad (2)$$

where  $x$  is a constant for a fixed position which depends on the fraction  $f$  of epithermal neutrons averaged over the foil position, and can be shown to be  $fE_c/4kT_0$ .  $G_t$  and  $G_r$  are the thermal and resonance self-shielding factors for the foils.

The activity under cadmium is

$$A_{\text{Cd}} = \sigma_0(g/\hat{R} - W'x + FG_r x I' / \sigma_0) \quad (3)$$

where  $F$  is the transmission of resonance neutrons through the cadmium covers,  $\hat{R}$  is the average cadmium ratio of a thin ' $1/v$ ' detector ( $v$  is neutron velocity) and  $W'$  is the small correction term of Walker *et al* (1960) which accounts for that part of a low-lying resonance which lies below  $E_{\text{Cd}}$ , the effective cadmium cut-off energy (see appendix 1).

The cadmium ratio, from equations (2) and (3), is

$$R = \frac{A}{A_{\text{Cd}}} = \left( \frac{gG_t + G_r x I' / \sigma_0}{g/\hat{R} - W'x + FG_r x I' / \sigma_0} \right). \quad (4)$$

In the experiment, the cadmium ratios of two standard foils, Au and Mn, are used to find the two unknown quantities  $x$  and  $1/\hat{R}$  in (4) assuming values for the other parameters (ie see § 3, tables 1 and 2). The value of  $I'/\sigma_0$  for any other isotope can then be calculated from its cadmium ratio using (4), together with suitable values for the other parameters.

### 3. The calculation of the self-shielding and $F$ factors

#### 3.1. Thermal self-shielding factor, $G_t$

The foils were all sufficiently thin, so that  $G_t$  was calculated from the simple formula given by Zweifel (1960) using a value of  $1.05\sigma_0$  for the effective thermal cross section to allow for spectral hardening.  $G_t$  factors for the foils varied between 1.00 and 0.96.

#### 3.2. Resonance self-shielding factor, $G_r$

The values of  $G_r$  for narrow mainly absorptive resonances, including Doppler broadening, have been calculated by Roe (1954). However, some of the Cu and Mn resonances are predominantly scattering resonances, and for Mn the calculations of Selander (1960) were used. To investigate the self-shielding factors for some of the scattering resonances of copper a Monte Carlo calculation was made. The problem is essentially one dimensional, the only spatial requirements being the depth of penetration into the foil where a scatter or capture occurs and the angles which the neutron tracks make with the normal to the surface of the foil. An isotropic  $1/E$  incident neutron flux was assumed, and foil edge effects were neglected. As a check it was shown that the results agreed, within the (about 1%) statistical uncertainty, with the calculations of Roe for Au and of Selander for Mn foils. It was shown that for the narrow copper resonances (which include appreciable scattering) the Roe calculations were accurate. For the wide  $^{63}\text{Cu}$  scattering resonance at 2060 eV, the unbroadened  $G_r$  value for a  $40 \text{ mg cm}^{-2}$  foil of natural Cu increased from 0.895 to  $0.932 \pm 0.013$ , but the resulting increase in  $G_r$  summed over all the resonances was only 0.2%. The two neighbouring resonances,  $^{63}\text{Cu}$  at 2660 eV and  $^{65}\text{Cu}$  at 2550 eV, cause interference, since neutrons scattered from the higher energy resonance can be captured in the lower energy resonance, slightly increasing its effective  $G_r$  value, while there is also a little shielding between the resonances which decreases  $G_r$ . An approximate calculation showed that the overall change in  $G_r$  was negligible. For  $^{63}\text{Cu}$  a small allowance was made for the unresolved resonances, with  $G_r$  factors of unity, using the calculations of Connolly *et al* (1968). In the case of  $^{65}\text{Cu}$  the  $G_r$  values for all the resolved resonances were greater than 0.95, and the effect of unresolved resonances was negligible. For  $^{107}\text{Ag}$ , Tb and Ho a sufficient number of resolved resonances were considered to calculate  $G_r$  accurately without making allowance for unresolved ones. The  $G_r$  values for the foils are given in table 3.

#### 3.3. $F$ factor

The transmission of epithermal neutrons through 1 mm cadmium,  $F$ , was taken as 0.991 for Au, since the principal resonance at 4.9 eV extends down into the cadmium cut-off region. Similarly with Tb and Ho, which have low-lying resonances at 2.35 and 3.92 eV, values of  $F$  were estimated to be 0.98 for these resonances, which produced effective  $F$  values when averaged over all the resonances of 0.995 and 0.99 respectively. In the case of Ho, neutron capture in the small fourth resonance at 18.2 eV was influenced by the nearby  $^{113}\text{Cd}$  resonance at 18.5 eV, but the overall effect on  $F$  was negligible. For Mn,  $^{63}\text{Cu}$ ,  $^{107}\text{Ag}$  and  $^{164}\text{Dy}$ ,  $F$  was taken as unity.

However for  $^{65}\text{Cu}$ , the narrow principal resonance at 229 eV is only 4.6 eV below the  $^{111}\text{Cd}$  resonance at 233.6 eV according to BNL 325 (1973). It is shown in appendix 2 that there is a small probability that neutrons which were resonance scattered in the

cadmium will enter the correct energy band to be resonantly captured in the copper. Thus there is enhanced resonance capture under cadmium, with an  $F$  value of 1.034 when averaged over the first five resonances.

#### 4. Measurements

##### 4.1. $\sigma_0$ of $^{159}\text{Tb}$ , $^{165}\text{Ho}$ and $^{107}\text{Ag}$

Pure foils of Tb and Ho were irradiated in the NPL standard thermal neutron flux, both bare and under 1 mm cadmium covers, and  $\sigma_0$  was found for the production of  $^{160}\text{Tb}$  (72 d half-life) and  $^{166g}\text{Ho}$  (27 h half-life) by the same activation method as described previously by Ryves (1970). The component uncertainties for these cross section measurements are given in table 2.

Table 1. Isotopic data.

Isotope	$g$ (20°C)	$F$	$W'$	Product half-life	Foil	
					Thickness (mg cm <sup>-2</sup> )	Composition
$^{55}\text{Mn}$	1	1	0	$2.580 \pm 0.002$ h <sup>b</sup>	15	Mn 88%—Ni 12%
$^{197}\text{Au}$	1.0053	0.991	0.066	$2.695 \pm 0.002$ d <sup>b</sup>	2–20	
$^{63}\text{Cu}$	1	1	0	$12.704 \pm 0.006$ h <sup>d</sup>	2–40	Natural Cu
$^{65}\text{Cu}$	1	1.034	0	$5.13 \pm 0.03$ min <sup>d</sup>	2–40	Natural Cu
$^{107}\text{Ag}$	1	1	0	$2.37 \pm 0.01$ min <sup>d</sup>	1–4	Natural Ag
$^{159}\text{Tb}$	1.0017	0.995	0.015	$71.93 \pm 0.2$ d <sup>c</sup>	16	
$^{164}\text{Dy}$	0.975 <sup>a</sup>	1	0	$139.2 \pm 0.2$ min <sup>b</sup>	30	Natural Dy 10%—Al 90%
$^{165}\text{Ho}$	1.0016	0.99	0.020	$26.83 \pm 0.03$ h <sup>d</sup>	20	

a Albert and Schumann (1967).

b Preferred half-life data (Lederer *et al* 1967).

c M J Woods (1973, private communication).

d Present measurement.

Table 2. Percentage uncertainties in  $\sigma_0$  for  $^{139}\text{Tb}$  and  $^{165}\text{Ho}$ .

Uncertainties	$^{159}\text{Tb}$	$^{165}\text{Ho}$
Random (99% confidence level)	1.2	1.2
Systematic		
Flux	0.7	0.7
$\beta$ counting efficiency	1.4	1.0
Half-life	0.3	—
Thermal self-shielding	—	0.5
Total (in quadrature)	2.0	1.8

Four Tb foils were counted and gave  $\sigma_0 = 23.2 \pm 0.5$  b. This result agrees very well with  $22 \pm 2$  b measured by Alstad *et al* (1967) but not with the value of Pomerance (1951) of  $46 \pm 4$  b.

Five Ho foils gave  $\sigma_0 = 61.2 \pm 1.1$  b, in very good agreement with the recent crystal spectrometer measurement of  $61 \pm 3$  b by Zimmerman *et al* (1967).

In the case of  $^{107}\text{Ag}$ ,  $\sigma_0$  had previously been measured by Ryves (1971) assuming a half-life of 2.39 min. However, using the newly measured half-life (see § 4.3) of 2.37 min, this cross section must be increased by 1.2%, since the silver samples had been counted about two half-lives after irradiation, giving  $\sigma_0 = 37.6 \pm 1.2$  b.

**Table 3.**  $G_r$  values at 20 °C.

Foil thickness (mg cm <sup>-2</sup> )	Mn-Ni	<sup>63</sup> Cu	<sup>65</sup> Cu	<sup>107</sup> Ag	Tb	<sup>164</sup> Dy	Ho	Au
5		0.962	0.992	0.961		1.000		0.737
10		0.932	0.986					0.613
15	0.900	0.909	0.982		0.835			0.532
20		0.889	0.978		0.802		0.700	0.486
25		0.870	0.974					
30		0.854	0.971					
35		0.839	0.967					
40		0.822	0.964					

#### 4.2. Resonance integrals by the cadmium ratio method

A slowing down epithermal neutron flux was obtained from the moderation of fast neutrons in water. The neutron source was provided by the  $^9\text{Be}(\text{D}, \text{n})^{10}\text{B}$  reaction, using a 100  $\mu\text{A}$  magnetically analysed beam of 3 MeV deuterons from the Van de Graaff accelerator striking a thick beryllium metal target. The target was positioned at the centre of a large cylindrical water tank (height 2.4 m, diameter 2 m). The thermalized neutron flux density in the water towards the edge of the tank was continuously monitored with a fission chamber and a boron-coated ionization chamber, and the target output was held constant to about 1% by a servo-system.

Eight circular foils were irradiated simultaneously in each measurement, the foils being positioned at equal distances on the circumference of a 14 cm diameter circle, encased in a thin perspex wheel which rotated in the horizontal plane at 150 rpm. This technique ensured that each foil of equal area had an equal average irradiation. The position of the centre of the wheel was variable from 15 cm above, and from 8 cm in front of, the target outwards to the edge of the tank. The dysprosium and manganese foils were smaller than the others (1 cm<sup>2</sup> to 4 cm<sup>2</sup>), but it was shown experimentally that at the relatively large distances between foils and target, the average flux and cadmium ratio for the small and large foils were the same. Several different positions of the wheel gave no significant difference in the measured resonance integrals, showing that the shape of the slowing down neutron spectrum did not alter with position in the moderator.

For an irradiation the wheel was loaded with matched pairs of standard Au and Mn foils and also two pairs of foils of the isotope under investigation. The pairs of foils were placed beside each other on the wheel, one foil in a thin aluminium box and one foil encased in 1 mm cadmium covers, with the cadmium covered foils spaced alternately round the circle. The Cd caused a thermal neutron flux depression in the bare foils of about 3%, but because of the symmetry it was the same for all foils and could be neglected. The measurement of each resonance integral depended only on

three sets of cadmium ratios, those of the two standard materials and the isotope concerned.

The activated foils were assayed in a  $4\pi\beta$  gas flow proportional counter making the usual corrections for dead-time and background. The relative masses and  $\beta$ -counting efficiencies of unmatched pairs of foils were measured directly by irradiating them side by side in the NPL standard thermal neutron flux facility and measuring the ratios of  $\beta$  activities produced. The small variation in  $\beta$  counting efficiency for cadmium covered and bare foils, which depends on the non-uniform neutron capture in the samples due to flux shielding effects, was negligible for the thin samples used.

#### 4.3. The half-lives of $^{64}\text{Cu}$ , $^{66}\text{Cu}$ , $^{108}\text{Ag}$ , $^{116g}\text{Ho}$ and $^{160}\text{Tb}$

The foils were irradiated several times, and the decaying activities measured in a  $4\pi\beta$  counter, and then analysed using a weighted least-squares program to determine the decay constants. The results obtained are given in table 1, where the uncertainties are random only, estimated at the 99% confidence level.  $^{64}\text{Cu}$  was counted over at least 5 half-lives and  $^{166g}\text{Ho}$  over 2.5 half-lives. The systematic uncertainties were estimated to be negligible compared with the random uncertainties; for example a change in the dead-time of 0.1  $\mu\text{s}$  would have changed the  $^{64}\text{Cu}$  half-life by only 0.0008 h. The activated Ho foils were checked carefully for any impurities by  $\gamma$  ray spectroscopy and no contamination was observed.

In the case of  $^{160}\text{Tb}$ , the half-life had been recently measured at NPL by M J Woods (1973, private communication) using an ionization chamber. Our irradiated foils contained a small contaminant with a  $\gamma$  spectrum and half-life corresponding to 139 min  $^{165}\text{Dy}$ , which was allowed to decay before counting the long-lived Tb.

## 5. Results and uncertainties

Results are given in table 4, together with some previous measurements and calculations from the literature. They are relative to the standards, Au:  $\sigma_0 = 98.8 \pm 0.3$  b;  $I' = 1514 \pm 60$  b and Mn:  $\sigma_0 = 13.23 \pm 0.2$  b;  $I' = 7.8 \pm 0.3$  b. The component uncertainties are listed in table 5.

The random uncertainties at a 99% confidence level were estimated from the dispersion of the measurements, and include the statistical counting uncertainties in the three cadmium ratios per measurement, together with uncertainties in the relative  $\beta$  efficiencies and masses of the foil pairs.

The greatest systematic uncertainty, due to the reference standards Au and Mn, is not easy to assess, since it depends mostly on the magnitudes of the cadmium ratios. This dependence has been investigated mathematically (see appendix 3) and it is found that if the uncertainties in the quantities  $(G_r I' / \sigma_0)$  for the two standards are both  $\pm 4\%$ , then the resulting uncertainties in  $(G_r I' / \sigma_0)$  for the measured isotopes are also approximately  $\pm 4\%$ , except for  $^{164}\text{Dy}$  where the error is much larger due to the peculiar variation of its capture cross section with energy.

The uncertainty in  $\sigma_0$  (especially for  $^{63}\text{Cu}$  and  $^{164}\text{Dy}$ ) contributes to the uncertainty in  $I'$ , which is consequently always greater than the uncertainty in  $(I' / \sigma_0)$ .

The value of  $(G_r I' / \sigma_0)$  is reasonably well known for the Au foils, but unfortunately in the case of Mn at the present time  $I'$  is not well established, our value of 7.8 b (selected from Axton 1963, Bardes *et al* 1962, Dahlberg *et al* 1961, Schmelz and Kohler 1967,

Table 4. Resonance integral results.

Isotope	Present measurements			Literature values			Comments
	$\sigma_0$ (b)	$I/\sigma_0$	$I$ (b)	$\sigma_0$ (b)	$I$ (b)	Reference	
$^{63}\text{Cu}$	$4.4 \pm 0.2$	$0.634 \pm 0.028$	$2.79 \pm 0.18$	$4.5 \pm 0.15$	$3.09 \pm 0.15$	Dahlberg <i>et al</i> (1961)	Beam geometry
				$4.5 \pm 0.15$	$3.17 \pm 0.18$	Baumann (1963)	—
				$4.3 \pm 0.2$	$3.7 \pm 0.5$	Borchardt (1967)	—
				4.5	2.4	Bennett (1961)	Re-evaluated by Baumann (1963)
				4.5	3.2	Anderson (1964)	Selected value; uncertain spectrum
$^{65}\text{Cu}$				$4.4 \pm 0.2$	$2.7 \pm 0.2$	Ryves (1970)	$I$ recalculated with $G_r = 0.88$
				—	3.31	BNL 325 (1973)	Calculated
				—	2.9	Simmons and McElroy (1970)	Calculated from new data
							— $1/v$ part, 1.7 b
$^{107}\text{Ag}$	$2.17 \pm 0.03$	$0.561 \pm 0.028$	$1.22 \pm 0.06$	$2.0 \pm 0.3$	$1.38 \pm 0.23$	Dahlberg <i>et al</i> (1961)	Beam geometry
				$2.0 \pm 0.3$	$1.39 \pm 0.22$	Baumann (1963)	—
				2.2	(1.1)	Bennett (1961)	Re-evaluated
				$2.17 \pm 0.3$	$1.17 \pm 0.12$	Ryves (1970)	—
				—	1.09	BNL 325 (1973)	Calculated. No allowance for unresolved resonances
$^{107}\text{Ag}$	$37.6 \pm 1.2$	$2.09 \pm 0.09$	$79 \pm 4$	29.5	61	Macklin and Pomerance (1956)	—
				$37.6 \pm 1.2$	$78 \pm 5$	Ryves (1971)	$\sigma_0$ increased, due to new half-life
				—	88	BNL 325 (1973)	Calculated



Table 4. (continued)

$^{159}\text{Tb}$	$23.2 \pm 0.5$	$16.8 \pm 1.0$	$390 \pm 24$	23.2	$380 \pm 40$	THAI-AEC-10 (1967)	Re-evaluated for $\sigma_0 = 23.2$ b — — $1/p$ part, 10 b. Reactivity method — Calculated using BNL 325
				$22 \pm 2$	$440 \pm 50$	Alstad <i>et al</i> (1967)	
				—	$620 \pm 25$	Scoville and Rogers (1968)	
				$22 \pm 2$	$343 \pm 35$	Ricabarra <i>et al</i> (1970)	
$^{164}\text{Dy}$	$2700 \pm 200$	$-0.19 \pm 0.04$	$-514 \pm 115$	2750	$390 \pm 30$	Walker (1969)	Total resonance activity above Cd cut-off energy Reactivity method Total resonance integral Calculated using BNL 325
				—	$(482 \pm 33)$	Jacks (1961)	
				—	$(317 \pm 34)$	Scoville and Fast (1964)	
$^{165}\text{Ho}$	$61.2 \pm 1.1$	$10.1 \pm 0.5$	$618 \pm 33$	64	38	Walker (1969)	No total error given — $1/p$ part, 30 b. Reactivity method includes formation of $^{166m}\text{Ho}$ Calculated, includes formation of $^{166m}\text{Ho}$
				67	$600 \pm 110$	THAI-AEC-10 (1967)	
				—	$830 \pm 25$	Scoville and Rogers (1968)	
				63	$685 \pm 30$	Walker (1969)	
			67	674		Stephenson (1967)	

**Table 5.** Percentage uncertainties in the resonance integrals.

Uncertainties	<sup>63</sup> Cu	<sup>65</sup> Cu	<sup>107</sup> Ag	<sup>159</sup> Tb	<sup>164</sup> Dy	<sup>165</sup> Ho
Random (at 99% confidence level)	1.4	1.9	1.4	3.7	6.8	2.9
Number of measurements	22	27	8	8	8	8
Systematic						
Au and Mn standards	4	4	4	4	20	4
$G_r$	1	—	—	1	—	1
$G_t$	—	—	—	—	1	—
$g$	—	—	—	—	1	—
$F$	—	2	—	1	—	1
$\sigma_0$	4.5	1.4	3.2	2.2	7.8	1.8
Total systematic (in quadrature)	6.1	4.7	5.1	4.8	21.5	4.6
Total (in quadrature)	6.3	5.1	5.3	6.1	22.5	5.4

Walker *et al* 1960) being somewhat lower than values in excess of 8 b which many other authors recommend. In appendix 3 the effect on the measured resonance integrals due to small changes in those of the standard materials has been calculated, so that if in the future more accurate values for the latter are obtained, our relative measurements can be simply adjusted. The coefficients  $\alpha'_1$  and  $\alpha'_2$  appropriate to our experimental conditions have been evaluated (table 6) so that  $\Delta I' = \alpha'_1 \Delta I'_{Au} + \alpha'_2 \Delta I'_{Mn}$ . It is seen that the resonance integrals of the Cu isotopes are especially sensitive to small fractional changes in  $I'_{Mn}$ , while Tb and Ho are influenced more by small fractional changes in  $I'_{Au}$ . The <sup>164</sup>Dy resonance integral is far more sensitive to small changes in the standard materials' parameters, than those of the other isotopes, due to the peculiar shape of its capture cross section, with a virtual resonance at  $-1.89$  eV, and its very small reduced resonance integral.

The present measurements of resonance integrals are more reliable than the previous ones by Ryves (1970) using the standard thermal neutron flux facility at the NPL because: (i) the slowing down neutron flux there was not  $1/E$ -dependent and large spectrum corrections, up to 20%, were necessary; and (ii) the epithermal flux fraction was much smaller, so that far less resonance activity was produced in the foils. The uncertainties quoted by many authors in table 4 have not been specified in enough

**Table 6.** Coefficients for computing the change in  $I'$  due to changes in  $I'_{Au}$ ,  $I'_{Mn}$ 

Isotope	$\alpha'_1$	$\alpha'_2$
<sup>63</sup> Cu	$2.6 \times 10^{-4}$	0.322
<sup>65</sup> Cu	$7.5 \times 10^{-5}$	0.144
<sup>107</sup> Ag	$4.21 \times 10^{-2}$	2.20
<sup>139</sup> Tb	$2.61 \times 10^{-1}$	-0.77
<sup>164</sup> Dy	-1.39	198
<sup>165</sup> Ho	$4.01 \times 10^{-1}$	1.22

detail to make a critical comparison of their results with ours very useful. It is probable that other authors have quoted their uncertainties at about a 68% confidence level, while ours are given at a 99% confidence level.

## 6. Discussion

### 6.1. $^{63}\text{Cu}$ and $^{65}\text{Cu}$ results

From the results in table 4 it can be seen that there is excellent agreement for  $^{63}\text{Cu}$  with the previous measurement by Ryves (1970), when the corrected foil resonance self-shielding factor is used in the latter. The accurate measurement of Dahlberg *et al* (1961) of  $3.09 \pm 0.15$  b is 11% higher than the present result of  $2.79 \pm 0.18$  b, but the two results are not directly comparable without some adjustments for the different values of  $\sigma_0$  and  $I'_{\text{Mn}}$  used. Normalizing to the same values of these parameters, a value of 4.4 b instead of 4.5 b for  $\sigma_0$ , and 7.8 b instead of their measured value of 8.15 b for  $I'_{\text{Mn}}$ , their result will be multiplied by  $\frac{4.4}{4.5} \times \frac{7.8}{8.15} = 0.936$  and become 2.89 b, in reasonable agreement. The other apparently accurate measurement, that of Baumann (1963), was made in a reactor, assuming a  $1/E$  flux, which in our experience is unlikely to be true. The measurement of Bennett (1961) is not described in detail, and large corrections are required as discussed by Baumann. The result of Anderson (1964) is selected from differing values derived from thick samples. The result of Borchardt (1967) has a somewhat greater quoted uncertainty than the other results. The resonance integral can also be calculated from the resonance parameters. Using data from BNL 325 (1973) a calculation gave 3.31 for the reduced resonance integral, but a recent evaluation by Simmons and McElroy (1970) using their own data yielded about 2.9 b, in reasonable agreement with the present measurement.

For  $^{65}\text{Cu}$  the present value of  $1.22 \pm 0.06$  b is in agreement with the previous measurement by Ryves (1970), but 13% below the value of Dahlberg *et al* (1961) of  $1.38 \pm 0.23$ . Again making some adjustments to their value for purpose of comparison,  $\sigma_0$  has to be changed from 2.0 to 2.17 b,  $I'_{\text{Mn}}$  from 8.15 to 7.8 b, and the  $F$  value from 1.0 to 1.034, giving a correction factor of  $\frac{2.17}{2.0} \times \frac{7.8}{8.15} \times \frac{1.0}{1.034} = 1.003$ . The net effect is to make no appreciable change to their result, and the large discrepancy remains. The Baumann result again assumes a  $1/E$  flux and agrees with the value of Dahlberg *et al* and the Bennett result has been adjusted very crudely to give about 1.1 b. The calculated resonance integral from BNL 325 (1973) resonance parameters is 1.09 b for the resolved resonances, which is broadly consistent with the present result.

### 6.2. $^{107}\text{Ag}$ results

There was excellent agreement with the previous measurement of Ryves (1971), after inserting the re-evaluated  $\sigma_0$ , but an approximate calculation using BNL 325 (1973) data gave a 10% larger result.

### 6.3. $^{159}\text{Tb}$ results

The value of  $I'$  was in reasonable agreement with THAI-AEC-10 (1967) and the calculation of Walker (1969), but higher than Ricabarra *et al* (1970) and lower than Alstad *et al* (1967), although just within the uncertainties. The value of Scoville and Rogers (1968),

using the pile reactivity method, is unaccountably high, which could possibly be due to some rare-earth impurity in their sample. Even for our 'spec-pure' samples, there was evidence of a slight dysprosium impurity, clearly seen in the activation analysis, but which was not detected by chemical or spectrographic analysis.

#### 6.4. $^{164}\text{Dy}$ results

The negative value of  $I'$  is due to the definition of reduced resonance integral adopted, and depends on the deviation of the thermal and epithermal shape of the neutron capture cross section from a  $1/\sqrt{E}$  dependence. The result is not inconsistent with the resonance integral measurements of Jacks (1961) and Scoville and Fast (1964). The value of  $\sigma_0$  is poorly known, and there was an inexplicably large spread in the eight measurements, which were all made under similar experimental conditions. The results were extremely sensitive to the reference standard parameters and to the spectrum, and this is reflected in the large 20% systematic uncertainty.

#### 6.5. $^{165}\text{Ho}$ results

The value obtained for  $I'$  was  $618 \pm 34$  b, which agrees well with the only other activation measurement in table 4, from THAI-AEC-10 (1967) of  $600 \pm 110$  b using the uncertainty of the associated cadmium ratio (17%). A pile reactivity experiment by Scoville and Rogers (1968) gave  $830 \pm 25$  b, which seems very high, although it also includes the resonance capture leading to the long-lived isomer  $^{166\text{m}}\text{Ho}$ . The reduced resonance integral  $I'$  has been calculated in some detail by Stephenson (1967) who obtained a value of 674 b (assuming  $\sigma_0 = 67$  b) and by Walker (1969) who obtained  $685 \pm 30$  b. Both these calculations include the production of  $^{166\text{m}}\text{Ho}$  as well as the 27 h  $^{166\text{g}}\text{Ho}$ , and are probably in agreement with our measurement.

#### 6.6. Conclusion

The present measurements of  $I'$  for  $^{63}\text{Cu}$ ,  $^{65}\text{Cu}$ ,  $^{107}\text{Ag}$ ,  $^{159}\text{Tb}$  and  $^{165}\text{Ho}$  all show improved agreement with calculations based on the resonance parameters, over most of the previous literature values. This helps to confirm the resonance data and establish a consistent set of resonance integrals.

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### Appendix 1. The calculation of $W'$

The resonance activity produced by neutrons between the effective cadmium cut-off energy  $E_{\text{Cd}}$  and the epithermal cut-off energy  $E_c$ , must be subtracted from the total

resonance activity in equation (3) by means of the quantity  $W'$  :

$$W' = \frac{1}{\sigma_0} \int_{E_c}^{E_{Cd}} \left[ \sigma_a(E) - g\sigma_0 \left( \frac{E_0}{E} \right)^{1/2} \right] \frac{dE}{E}. \quad (\text{A1.1})$$

It is assumed that only the lowest principal resonance departs from a  $1/\sqrt{E}$  energy dependence between the limits of integration. Using the single level Breit-Wigner formula for this resonance, the integral can be evaluated approximately if the total resonance width  $\Gamma \ll 2(E_{res} - E)$  where  $E_{res}$  is the resonant energy. Then

$$W' \simeq \left( \frac{E_0}{E_{res}} \right)^{1/2} \left| \frac{\sigma_{01}}{\sigma_0} (4\sqrt{x} + 2x\sqrt{x}) + (g-1)(2/\sqrt{x}) \right|_{x_1}^{x_2} \quad (\text{A1.2})$$

where  $\sigma_{01}$  is the value of that part of the thermal capture cross section due to the principal resonance at the energy  $E_0$  of 0.0253 eV, and  $x_2 = E_{Cd}/E_{res}$ ,  $x_1 = E_c/E_{res}$ . Exact integrations indicated that (A1.2) is accurate to better than 10%, which was sufficient for the present measurements.

The values of  $W'$  for Au, Tb and Ho were calculated using (A1.2) and are given in table 1, assuming  $E_c = 0.1$  eV,  $E_{Cd} = 0.677$  eV,  $T = 20^\circ\text{C}$ .

In (A1.1) the factors for resonance self shielding  $G_r$ , and the transmission of resonance neutrons through cadmium  $F$ , have been omitted since they are both close to unity, and it is assumed that  $W' = W'G_rF$ .

## Appendix 2. The effective $F$ for $^{65}\text{Cu}$

The cadmium ratio of  $^{65}\text{Cu}$  is influenced by the proximity of a prominent resonance in cadmium to the principal low energy copper resonance. The resonance in  $^{111}\text{Cd}$  at 233.6 eV is sufficiently close to the  $^{65}\text{Cu}$  resonance at 229 eV for a few neutrons which are resonance-scattered in the cadmium to lose sufficient energy to enhance the copper resonance absorption. The energy loss of elastically scattered neutrons from cadmium at this energy has a rectangular distribution between 0 and 8.3 eV. The effect is small because the resonance energy widths in question are both narrow, and the  $^{111}\text{Cd}$  isotopic abundance is only 12.86%, but cannot be neglected, for the 1 mm thick cadmium covers used, since the probability of resonance scattering is high.

The enhanced activity in  $^{65}\text{Cu}$  can be calculated approximately, assuming that the plane cadmium covers and copper foil in the sandwich are infinite in extent, ignoring the edge effects.

Let the foil thicknesses be  $t$ , number of nuclei per cubic centimetre be  $N$ , the cross sections be  $\sigma$ , with the appropriate suffices Cd or Cu for the isotopic materials and  $s$ ,  $a$  for scattering or absorption.

An isotropic slowing down flux  $\phi(E) = k/E$  per unit energy  $E$  is incident in the sample (where  $k$  is a constant), and neutrons are elastically resonance-scattered in the  $^{111}\text{Cd}$  and resonance-absorbed in the  $^{65}\text{Cu}$ . Consider the number of neutrons scattered in an area  $S$  cm<sup>2</sup> of the cadmium (including both sides of the sandwich) which subsequently pass through the copper foil. It is assumed from a consideration of the symmetry that half the total number of these scattered neutrons pass through the copper foil, while half escape outwards missing the copper. The scattered neutrons are assumed to be also isotropic, with average energies equally distributed between  $E$  and  $\alpha_{Cd}E$  where  $\alpha_{Cd} = [(1-A)/(1+A)]^2$  and  $A$  is the atomic number of  $^{111}\text{Cd}$ . (In fact those neutrons

scattered forward from the first cadmium sheet have higher energies than those back-scattered from the second cadmium sheet, but the average energy distribution of all scattered neutrons in the copper is assumed to have this form.)

Then the number of cadmium-scattered neutrons of incident energy  $E$  to  $E + dE$  which are captured in copper between energies  $E'$  to  $E' + dE'$  is

$$St_{\text{Cd}}G_{\text{rCd}}(E)\phi(E)N_{\text{Cd}}\sigma_{\text{sCd}}(E) dE \frac{P(E') dE'}{E - \alpha_{\text{Cd}}E} \quad (\text{A2.1})$$

where  $G_{\text{r}}(E)$  is the resonance self-shielding factor and  $P(E')$  is the probability of capture in copper:

$$P(E') = G_{\text{rCu}}(E')N_{\text{Cu}}\sigma_{\text{aCu}}(E')l \quad (\text{A2.2})$$

where  $l$  is the average track length through the copper foil,  $2t_{\text{Cu}}$  for an isotropic flux.

Expression (A2.1) can be approximately integrated over the narrow non-overlapping resonances of  $^{111}\text{Cd}$  at  $E$  and  $^{65}\text{Cu}$  at  $E'$  in terms of the resonance integrals  $I'$  and averaged self-shielding factors  $G_{\text{r}}$ , where

$$G_{\text{r}}I' = \int_{\text{resonance}} G_{\text{r}}(E)\sigma(E) dE/E$$

with appropriate suffices, making the good assumption that the resonance widths are very much less than  $E$ . This implies that one can write

$$G_{\text{r}}I' \simeq \frac{1}{E} \int_{\text{resonance}} G_{\text{r}}(E)\sigma(E) dE$$

with sufficient accuracy for these narrow resonances, and hence in (A2.1) the denominator can be treated as a constant, and also that one can replace  $(E - \alpha_{\text{Cd}}E)$  by  $(E' - \alpha_{\text{Cd}}E')$  since  $E \simeq E'$ .

Thus on integration (A2.1) becomes

$$2kSt_{\text{Cd}}t_{\text{Cu}}N_{\text{Cd}}N_{\text{Cu}}G_{\text{rCd}}G_{\text{rCu}}I'_{\text{aCu}}I'_{\text{sCd}}/(1 - \alpha_{\text{Cd}}). \quad (\text{A2.3})$$

The direct resonance activation of the copper foil is

$$kSt_{\text{Cu}}N_{\text{Cu}}G_{\text{rCu}}I'_{\text{aCu}}. \quad (\text{A2.4})$$

Thus the resonance activity of the bare copper foil is increased with the cadmium covers by a factor

$$F_1 = 1 + \frac{2t_{\text{Cd}}N_{\text{Cd}}G_{\text{rCd}}I'_{\text{sCd}}}{1 - \alpha_{\text{Cd}}} \quad (\text{A2.5})$$

(where the suffix 1 refers to this first resonance). Inserting numerical values,  $t_{\text{Cd}} = 0.1$  cm,  $(1 - \alpha_{\text{Cd}}) = 0.0354$ ,  $I'_{\text{sCd}} = 2.51$  b,  $N_{\text{Cd}} = 5.95 \times 10^{21}$  atoms  $^{111}\text{Cd}$  cm $^{-3}$  and  $G_{\text{r}} \simeq 0.50$  for 0.2 cm total thickness of Cd (including Doppler broadening) and hence  $F_1 = 1.042$  for this resonance.

The  $F_i$  values for the other  $i$  resonances of  $^{65}\text{Cu}$  are close to unity, and the average overall  $F$  value for  $^{65}\text{Cu}$  is found by summing their resonances (suffix  $i$ ).

$$F = \sum_i F_i I'_i / I'$$

**Appendix 3. The systematic uncertainty due to the reference standards**

An expression is derived from equation (4) by straightforward algebraic manipulation which relates the resonance integrals of the reference standards and isotope being measured, of the form

$$Q = \alpha_1 Q_{\text{Au}} + \alpha_2 Q_{\text{Mn}} \tag{A3.1}$$

where  $Q = (G_r I' / \sigma_0)$  and  $\alpha$ -suffices 1 and 2 refer to Au and Mn respectively. For simplicity it is assumed that  $W' = 0$  and  $g = 1$ . Then

$$\alpha_1 = \frac{(F' - 1/R)_1 (G_1/R - G_{12}/R_2)}{(F' - 1/R)(G_{11}/R_1 - G_{12}/R_2)} \tag{A3.2}$$

and  $\alpha_2$  is a similar expression, with suffices 1 and 2 reversed.  $F' \equiv FG_r(\text{Cd})/G_r(\text{bare})$  if the two irradiated foils, bare and under cadmium, are not perfectly matched. In the equations  $G_r$  refers to the bare foils.

The fractional change in  $Q$  due to changes in  $Q_{\text{Au}}$  and  $Q_{\text{Mn}}$  is

$$\frac{\Delta Q}{Q} = \frac{\alpha_1 \Delta Q_{\text{Au}} + \alpha_2 \Delta Q_{\text{Mn}}}{\alpha_1 Q_{\text{Au}} + \alpha_2 Q_{\text{Mn}}} \tag{A3.3}$$

If equal independent fractional uncertainties are assumed, so that  $\Delta Q_{\text{Au}} = \pm \epsilon Q_{\text{Au}}$ ,  $\Delta Q_{\text{Mn}} = \pm \epsilon Q_{\text{Mn}}$ , the fractional uncertainty in  $Q$ ,  $|\Delta Q/Q| \leq \epsilon$  if  $\alpha_1 \geq 0$ ,  $\alpha_2 \geq 0$ , ie if both terms in the denominator of (A3.3) are positive. These conditions can be very simply interpreted physically from the expressions for  $\alpha_1$  and  $\alpha_2$  in (A3.2), since in the present experiments,  $G_1 \simeq 1$ ,  $F' \gg 1/R$  and  $R_{\text{Mn}} \gg R_{\text{Au}}$ , as

$$\frac{1}{R} - \frac{1}{R_{\text{Mn}}} \geq 0 \quad \frac{1}{R_{\text{Au}}} - \frac{1}{R} \geq 0,$$

that is,

$$R_{\text{Mn}} \geq R \geq R_{\text{Au}} \tag{A3.4}$$

If the fractional uncertainties of  $Q_{\text{Au}}$  and  $Q_{\text{Mn}}$  are both  $\pm 4\%$ , then that of  $Q$  for the isotope being measured is  $4\%$  or less if (A3.4) holds, which is true for all the isotopes except  $^{159}\text{Tb}$  (where  $\alpha_2$  is only slightly negative) and  $^{164}\text{Dy}$ . However, since the values of  $I'_{\text{Au}}$  and  $I'_{\text{Mn}}$  are not entirely independent, due to the normal methods of measurement of  $I'_{\text{Mn}}$  relative to the primary standard  $I'_{\text{Au}}$  reported in the literature, the upper limit of  $\pm 4\%$  has been taken as the systematic uncertainty in all cases, except  $^{164}\text{Dy}$ . For  $^{164}\text{Dy}$ ,  $\alpha_1 < 0$ ,  $\alpha_2 > 0$  and the decrease in the denominator of (A3.3) significantly increases the uncertainty to  $\pm 20\%$ .

The effect on a measured resonance integral of small changes in the resonance integrals of the standards is easily derived from equation (A3.1) as

$$\Delta I' = \alpha'_1 \Delta I'_{\text{Au}} + \alpha'_2 \Delta I'_{\text{Mn}} \tag{A3.5}$$

where

$$\alpha'_i = \alpha_i \frac{G_{ri} \sigma_0}{G_r \sigma_{0i}} \quad (i = 1, 2).$$

Average numerical values of  $\alpha'_1$  and  $\alpha'_2$  are given in table 6 for the particular experimental conditions. As an example, consider the effect on  $I'_{Ag}$  of +4% changes in  $I'_{Au}$  and  $I'_{Mn}$ , ie  $\Delta I'_{Au} = 60.6$  b and  $\Delta I'_{Mn} = 0.312$  b. Then from (A3.5) and table 6,

$$\Delta I'_{Ag} = (4.21 \times 10^{-2})(60.6) + (2.20)(0.312) = 3.24 \text{ b,}$$

an increase of 4.1% in  $I'_{Ag}$ .

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