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THE (n, γ) CROSS SECTION OF ^{197}Au AT 30 AND 64 keV NEUTRON ENERGY

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Abstract—The neutron capture cross section of ^{197}Au was measured at 30 and 64 keV neutron energy using the activation method. Two different methods were used at 30 keV for the absolute neutron flux determination. The results are $\sigma_{n,\gamma}^{Au}$ (30 keV) = (0·600 ± 0·009) barn and $\sigma_{n,\gamma}^{Au}$ (64 keV) = (0·357 ± 0·009) barn.

1. INTRODUCTION

THE neutron capture cross sections in the keV energy region are of considerable interest to nuclear theory in connexion with the optical model (LANE and LYNN, 1957), to cosmological theories of element formation (BURBIDGE *et al.*, 1957) and to the design of nuclear reactors. These important applications have led to a large number of experiments by a variety of techniques which for the determination of reaction rates fall mainly into two types: (i) The detection of prompt γ-rays by a large liquid scintillator tank (DIVEN *et al.*, 1960; GIBBONS *et al.*, 1961), a Moxon–Rae detector (MOXON and RAE, 1963) or a proportional counter (KONKS *et al.*, 1964). (ii) The detection of the radioactivity which is induced by neutron capture in many nuclei (MACKLIN *et al.*, 1957; BOOTH *et al.*, 1958). However, the main problem seems to be the absolute neutron flux strength determination. Only the shell transmission method (SCHMITT, 1963; BELANOVA, 1960) avoids any absolute measurements but the corrections for the resonance self-shielding and for secondary neutron scattering effects are considerable.

The (n, γ) cross section of gold is of special interest as a standard. Existing measurements are shown in Fig. 1. The cross-section values measured by different groups disagree by a factor of about two.

This measurement of the neutron capture cross section of gold is presented mainly to give an absolute value at 30 keV for the renormalization of energy-dependent cross sections measured by other authors. To avoid systematic errors two different methods were used for the determination of $\sigma_{n,\gamma}^{Au}$ at 30 keV neutron energy.

2. EXPERIMENTAL PROCEDURE AND RESULTS

Neutrons for the irradiation of the gold samples were obtained from the reactions ^{7}Li(p, n)^{7}Be and ^{3}H(p, n)^{3}He. The protons were accelerated by a 3 MeV Van de Graaff. A layer of LiF evaporated on a 0·06-cm thick Cu backing plate was used as the target at 30 keV neutron energy. Just above the thresholds for the ^{7}Li(p, n)^{7}Be and ^{3}H(p, n)^{3}He reactions one gets neutrons of 30 and 64 keV average neutron energy. In this case the neutrons are kinematically collimated in a forward cone. Therefore it was possible to use a water-cooling ring around the edge of the Cu backing plate which did not interfere with the neutron flux. This method enabled us to bombard the target with a proton beam current of about 150 μA.

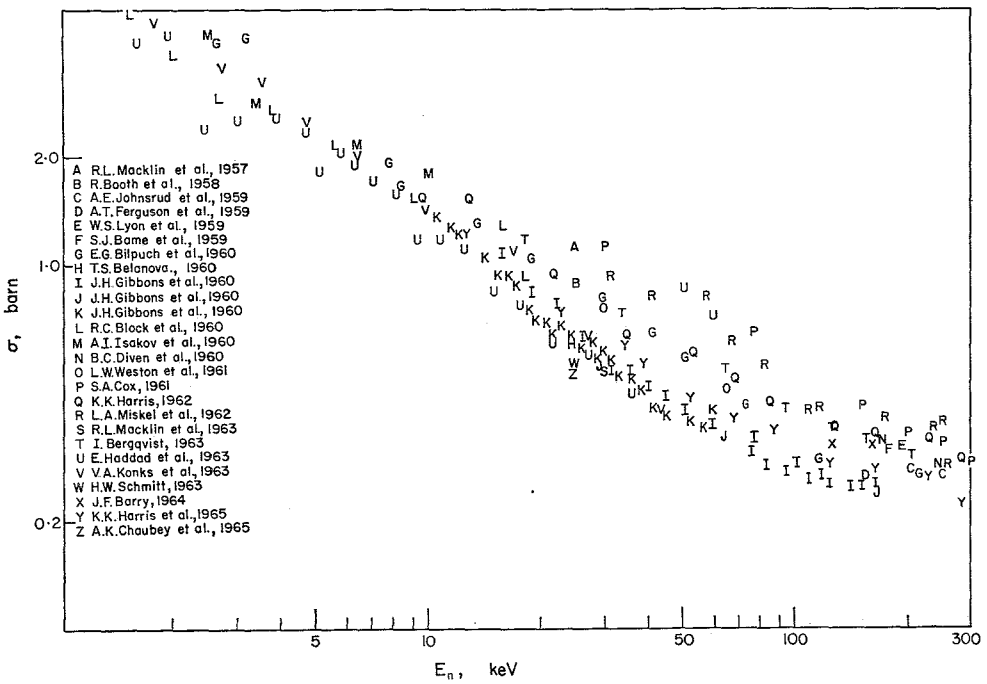

FIG. 1.—The (n, γ) cross section of ^{197}Au. Comparison of existing measurements.

The proton beam energy was measured and controlled by a proton spin resonance spectrometer. It was calibrated by the threshold energies of the ^{7}Li(p, n)^{7}Be and ^{3}H(p, n)^{3}He reactions and the opening angle of the forward cone of the neutrons from the ^{7}Li(p, n)^{7}Be reaction. This cone was measured using long counters. The uncertainty in the primary energy is about 1 keV.

The Au samples were 0·03-cm thick foils. In this case we have $\bar{\sigma}_{\mathrm{tot}} \,.\, N \,.\, d \ll 1$ and the cross section can be determined by its definition equation

$$C = \sigma \,.\, N \,.\, d \,.\, Q \tag{1}$$

where C is the reaction rate (activation) per sec . cm^2; Q is the neutron flux strength per sec . cm^2; N is the number of atoms per cm^3 and d is the thickness of the foil. However, for the application of equation (1) many corrections must be taken into consideration.

2.1 *The cross section at* 30 *keV* (1*st method*)

The principle of this method is to determine the neutron flux strength by the radioactivity of the reaction product ^{7}Be in the LiF target (PERRY, 1960). In this case, it is of special advantage, that the γ-ray energies occurring in the decays of ^{7}Be and ^{198}Au are very similar and therefore the ratio of the γ-efficiencies of the used NaI(Tl) detector is near to one.

The experimental set-up is shown in Fig. 2. The proton beam is limited by an aperture to 0·9 cm diameter and strikes a LiF target. On the LiF a thin layer of Al is evaporated to avoid any loss of ^{7}Be. The gold sample of 1·8-cm diameter is exposed to the neutron flux at the back of the Cu target backing. A second 0·005-cm thick

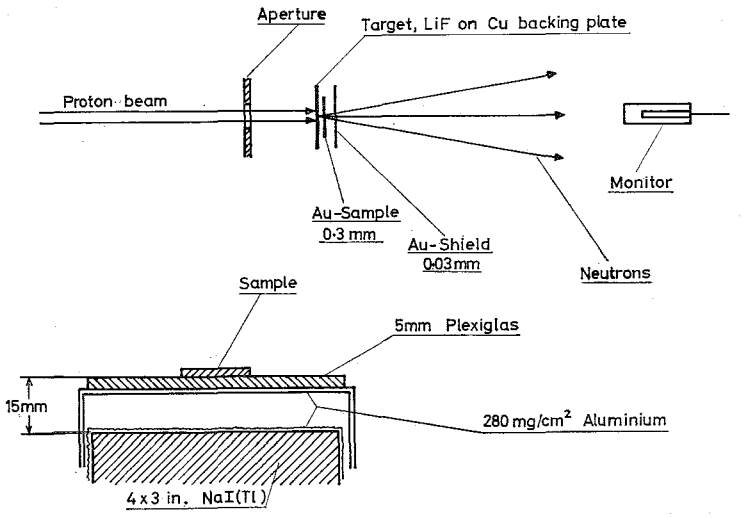

FIG. 2.—Experimental set-up. (First method).

gold foil is exposed to shield the sample from activation by thermal and electron volt resonance neutrons scattered back from the room. The activity of the external circle of this foil, which was not struck by the kinematically collimated neutrons was measured after the irradiation. No measurable activity was found showing that activation due to scattered neutrons was negligible.

Two different experiments were carried out using 15 and 50-min irradiation times. The time dependence of the neutron flux due to intensity variation of the proton beam current was monitored by a long counter 2 m distant from the target. After the irradiations the γ-activities of the target (^{7}Be; 438 keV transition of ^{7*}Li) and of the sample (^{198}Au; 411 keV transition of ^{198*}Hg) were measured in the same geometric arrangement using a 4 × 3 in. NaI(Tl) detector. The γ-ray spectra were analysed by a multichannel analyser. The experimental points of the photopeaks were fitted by gaussian curves.

The quantitative connexion between the counts Z in the photopeak and the activation C is easy to get from the differential equations of the radiative decay

$$\begin{aligned} Z_{\mathrm{Au}} &= \epsilon_{\mathrm{Au}} \cdot C_{\mathrm{Au}} \cdot F_{\mathrm{Au}} \\ Z_{\mathrm{Be}} &= \epsilon_{\mathrm{Be}} \cdot Q \cdot F_{\mathrm{Be}} \\ F &= \tfrac{1}{\lambda} \cdot (1 - \mathrm{e}^{-\lambda T}) \cdot (1 - \mathrm{e}^{-\lambda\theta}) \cdot \mathrm{e}^{-\lambda t} \cdot T_M. \end{aligned} \tag{2}$$

ϵ is the probability that a count occurs in the photopeak when a decay takes place. The factor F depends on the decay constant λ, the irradiation time T, the counting time θ, the time t between the end of the irradiation and the beginning of the counting and on the correction factor T_M calculated from the monitor counting rates. The factors

$$\begin{aligned} \epsilon_{\mathrm{Au}} &= \frac{a_{\mathrm{Au}}}{1 + \alpha_{\mathrm{Au}}} \cdot \eta_{\mathrm{Au}} \cdot P_{\mathrm{Au}} \cdot h_{\mathrm{Au}}{}' \cdot h_{\mathrm{Au}} \\ \epsilon_{\mathrm{Be}} &= a_{\mathrm{Be}} \cdot \eta_{\mathrm{Be}} \cdot P_{\mathrm{Be}} \cdot h_{\mathrm{Be}}{}' \end{aligned} \tag{3}$$

contain the transition intensity per decay a for the transition considered. α is the internal conversion coefficient, η the total detection efficiency of the NaI(Tl) crystal and P its peak-to-total ratio. The factors h' and h represent the γ-ray absorption in the cover of the NaI(Tl) detector and the γ-ray self-absorption in the Au sample.

η and P were evaluated using the calculated values of WEITKAMP (1963) and HEATH (1964). The γ-absorption between the γ-source and the NaI(Tl) crystal was calculated using the formula

$$h' = \mathrm{e}^{-\mu\delta} \tag{4}$$

for γ-rays crossing the cover perpendicular to the crystal surface. μ is the total absorption coefficient and δ the thickness of the absorption material. Values of μ were taken from GRODSTEIN (1956). It should be possible to get precise values of the factor

$$W = \frac{\eta_{\mathrm{Be}} \cdot P_{\mathrm{Be}} \cdot h_{\mathrm{Be}}'}{\eta_{\mathrm{Au}} \cdot P_{\mathrm{Au}} \cdot h_{\mathrm{Au}}'} \tag{5}$$

because of the similar γ-ray energies.

The γ-ray self-absorption in the Au sample was calculated using the assumption of homogenous activation. This leads to the formula

$$h = \frac{1}{\mu\delta} \cdot \frac{\int_{\Omega} \int_{0}^{\mu\delta} \exp\left(-\frac{\mu x}{\cos\theta}\right) \mathrm{d}\mu x \,.\, \mathrm{d}\Omega}{\int_{\Omega} \mathrm{d}\Omega} \,. \tag{6}$$

The integration limit Ω is given by the maximum opening angle of the γ-rays measured by the NaI(Tl) detector.

The activation C_{Au} contains the cross section

$$C_{\mathrm{Au}} = Q \,.\, N \,.\, d_{\mathrm{Au}} \cdot g(d_{\mathrm{Cu}}) \cdot f(d_{\mathrm{Au}}) \,.\, S(d_{\mathrm{Au}}) \,.\, \sigma_{\mathrm{n},\gamma}^{\mathrm{Au}}. \tag{7}$$

The factor $g(d_{\mathrm{Cu}})$ takes into account the loss of neutrons in the Cu target backing plate and the factor $f(d_{\mathrm{Au}})$ the correction for the longer neutron flight path in the sample due to the divergence of the kinematically collimated beam. Values were calculated using

$$g(d_{\mathrm{Cu}}) = \frac{\int_{\cos\theta}^{1} \exp\left(-\frac{\Sigma_{\mathrm{tot}}^{\mathrm{Cu}} d_{\mathrm{Cu}}}{\cos\theta}\right) d\cos\theta}{\int_{\cos\theta}^{1} d\cos\theta} \tag{8}$$

and

$$f(d_{\mathrm{Au}}) = \frac{1}{\Sigma_{\mathrm{tot}}^{\mathrm{Au}} \cdot d_{\mathrm{Au}}} \cdot \frac{\int_{\cos\theta}^{1} \int_{0}^{\Sigma_{\mathrm{tot}}^{\mathrm{Au}} d_{\mathrm{Au}}} \exp\left(-\frac{\Sigma_{\mathrm{tot}}^{\mathrm{Au}} X}{\cos\theta}\right) \mathrm{d}\Sigma_{\mathrm{tot}}^{\mathrm{Au}} X \,.\, d\cos\theta}{\int_{\cos\theta}^{1} d\cos\theta} \,. \tag{9}$$

Here θ is the maxima opening angle of the neutron cone and Σ_{tot} the total macroscopic cross section. The factor $S(d_{\mathrm{Au}})$ includes the resonance self-shielding of the foil (DRESNER, 1962) and the activation by neutrons which are scattered within the

TABLE 1(a).—VALUES NEEDED FOR CROSS-SECTION CALCULATION

Notation	Value	Remarks
a_{Au}	0·998	*Nuclear Data Sheets* (1958)
α_{Au}	0·042	LEWIN (1963), PETTERSON (1965)
a_{Be}	0·1032	TAYLER and MERRITT (1962)
η_{Au}	0·2674	Interpolated between values calculated by WEITKAMP and HEATH
P_{Au}	0·752	
η_{Be}	0·2551	
P_{Be}	0·702	
h_{Au}'	0·911	Calculated using equation (4)
h_{Be}'	0·916	
h_{Au}	0·930	using equation (6)
$g(d_{Cu})$	0·975	using equation (8)
$f(d_{Au})$	0·999	using equation (9)
$S(d_{Au})$	1·021	MACKLIN

TABLE 1(b).—MEASURED VALUES FOR THE CROSS-SECTION CALCULATION AT 30 keV NEUTRON ENERGY

Notation	Values	
Measurement	No. 1	No. 2
Z_{Au}	2·214 10^6	1·346 10^5
Z_{Be}	1·028 10^6	1·500 10^5
F_{Be}/F_{Au}	4·914 10^{-3}	1·141 10^{-2}
Au weight	1·533 g	1·463 g
$\sigma_{n,\gamma}$	0·5913 barn	0·5942 barn

TABLE 1(c).—ESTIMATED ERRORS OF THE VARIOUS VALUES USED FOR THE CROSS-SECTION CALCULATION

Notation	Error (%)	Notation	Error (%)
a_{Au}	0·1	Z_{Au}/Z_{Be}	0·6
$1 + \alpha_{Au}$	0·2	F_{Be}/F_{Au}	0·4
a_{Be}	1·6	h_{Au}	0·5
W	0·8	$S(d_{Au})$	0·5
$g(d_{Cu})$	0·3	$N \cdot d_{Au}$	0·1

foil (SCHMITT, 1960) and was calculated using the simplified relations for $S(d)$ of MACKLIN.

Table 1(a) contains all values which are needed for the calculation of the cross section. Table 1(b) contains the measured values and Table 1(c) the errors of the values given in Tables 1(a) and (b). The result for the cross section is

$$\sigma_{n,\gamma}^{Au}(30\text{ keV}) = (0{\cdot}593 \pm 0{\cdot}012)\text{barn}. \qquad (10)$$

The error in this first method is mainly due to the error of a_{Be}.

2.2 *The cross section at* 30 *keV and* 64 *keV* (*2nd method*)

The principle of this method is to determine the neutron flux by the manganese bath method (PERRY). The radioactivity of the Au sample was determined absolutely by the coincidence method.

The experimental set-up is shown in Fig. 3. The kinematically collimated neutrons entered a glass sphere through an entrance channel made of quartz. The neutrons slowed down in the $MnSO_4$ solution and were captured at thermal energies by the various components of the manganese bath. The integration over the space-dependent neutron flux was performed after the irradiation by stirring the solution. A well-defined quantity of the solution was filled into a container of Plexiglas and its activity was measured using a 4 × 3 in. NaI(Tl) crystal detector. The γ-activity of the Au

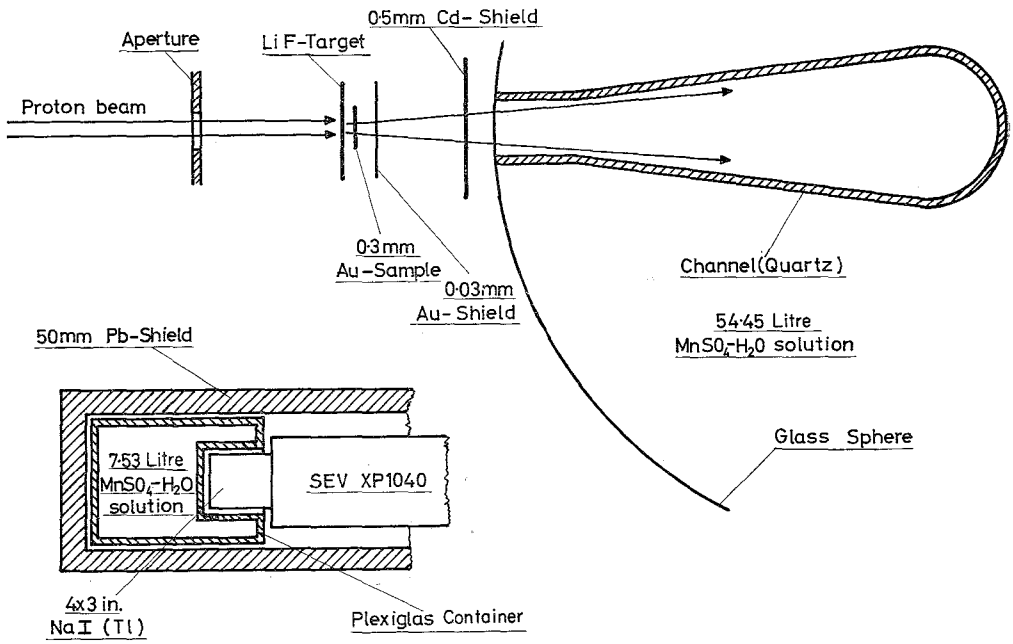

FIG. 3.—Experimental set-up (Second method).

sample was measured also by the NaI(Tl) detector. The counts in the photopeaks are given by the following relations:

$$Z_{Au} = \epsilon_{Au} \cdot Q \cdot N \cdot d_{Au} \cdot F_{Au} \cdot g_{Au}(d_{Cu}) \cdot f(d_{Au}) \cdot S(d_{Au}) \cdot \sigma_{n,\gamma}^{Au}$$
$$Z_{Mn} = \epsilon_{Mn} \cdot C_{Mn} \cdot F_{Mn} \cdot g_{Au}(d_{Cu}) \cdot g_{Mn}(d_{Au}, d_{Cd}). \qquad (11)$$

The meaning of all factors is the same as in Section 2.1.

The relations C_{Mn}/Q needed for cross-section calculation follows from a few well-known simple considerations: The 'source' strength of the neutron beam is

$$Q = \int_0^\infty \int_V [\Sigma_H(E) + \Sigma_0(E) + \Sigma_S(E) + \Sigma_{Mn}(E)] \cdot \phi(E, \mathbf{r})\, dV\, dE + Q_L + Q_C. \qquad (12)$$

$\Sigma(E)$ is the macroscopic absorption cross section at the energy E, $\phi(E, \mathbf{r})$ is the neutron flux in the manganese bath solution at the point $\mathbf{r}$ and V is the volume of the sphere. Q_L is the loss of neutrons due to the leakage at the edge of the sphere and Q_C is the loss of neutrons due to the capture in the entrance channel. Using the well-known

thermal cross sections (BECKURTS and WIRTZ, 1964) or their ratios (AXTON *et al.*, 1965) and regarding the resonance contribution of the Mn activation one gets

$$\frac{C_{\mathrm{Mn}}}{Q} = \frac{1{\cdot}011 - Q_L/Q - Q_C/Q}{1{\cdot}040 + 0{\cdot}02497\,.\,N_H/N_{\mathrm{Mn}}}\,. \tag{13}$$

Q_C/Q was calculated using thermal neutron flux values measured by gold foils in the neighbourhood of a Sb–Be source in the solution. Q_L/Q was determined using relative long counter measurements and absolute neutron flux values measured by gold foils in the solution near the edge of the glass sphere.

ϵ_{Au} and ϵ_{Mn} were determined using samples of known absolute disintegration rates measured by the $4\pi\beta$–γ coincidence apparatus (CAMPION, 1958). The determination of the absolute activity of the thick Au foils was described in an earlier paper (PÖNITZ, 1963). To get a well-known ^{56}Mn activity $MnSO_4$ powder was activated by thermal neutrons and its activity determined by the $4\pi\beta$–γ coincidence method. To avoid errors an extrapolation method was used (PÖNITZ and PÖNITZ and BRUDERMÜLLER, 1963).

Tables 2(a), (b), (c) contain the factors needed for the cross-section calculation,

TABLE 2(a).—VALUES NEEDED FOR CROSS-SECTION CALCULATION

Notation	Value	Remarks
ϵ_{Au}	0·1922	$4\pi\beta$–γ coincidence
ϵ_{Mn}	$0{\cdot}969 \times 10^{-3}$	$4\pi\beta$–γ coincidence
$g_{\mathrm{Mn}}(d_{\mathrm{Au}}, d_{\mathrm{Cd}})$	0·954	Calculated using equation (8)
$S(d_{\mathrm{Au}})$, (30 keV)	1·021	MACKLIN
$S(d_{\mathrm{Au}})$, (64 keV)	1·023	MACKLIN
Q_L/Q	0·069	Measured (see text)
Q_C/Q	0·005	Measured (see text)

TABLE 2(b).—MEASURED VALUES FOR THE CROSS-SECTION CALCULATION

Notation	Values	
E_n	30 keV	64 keV
Z_{Au}	$4{\cdot}378 \times 10^5$	$1{\cdot}403 \times 10^5$
Z_{Mn}	$6{\cdot}991 \times 10^5$	$2{\cdot}606 \times 10^5$
$F_{\mathrm{Au}}/F_{\mathrm{Mn}}$	0·940	1·422
Au weight	1·465 g	1·468 g
$\sigma_{\mathrm{n},\gamma}$	0·6037 barn	0·360 barn

TABLE 2(c).—ESTIMATED ERRORS OF THE VARIOUS VALUES GIVEN IN TABLES 2(a) AND (b)

Notation	Error (%)	Notation	Error (%)
ϵ_{Au}	0·8	$1 + Q_C/Q$	0·2
ϵ_{Mn}	0·8	$Z_{\mathrm{Au}}/Z_{\mathrm{Mn}}$	0·6
$g_{\mathrm{Mn}}(d_{\mathrm{Au}}, d_{\mathrm{Cd}})$	0·4	$F_{\mathrm{Au}}/F_{\mathrm{Mn}}$	0·5
$S(d_{\mathrm{Au}})$	0·5	$N\,.\,d_{\mathrm{Au}}$	0·1
$1 + Q_L/Q$	1·3	—	—

the measured values and its errors. The result using a LiF target ($E_n = 30$ keV) is

$$\sigma_{n,\gamma}^{Au}(30\text{ keV}) = (0{\cdot}604 \pm 0{\cdot}011)\text{barn} \tag{14}$$

and using a ZrH^3 target ($E_n = 64$ keV) is

$$\sigma_{n,\gamma}^{Au}(64\text{ keV}) = (0{\cdot}360 \pm 0{\cdot}008)\text{barn}. \tag{15}$$

For this method the main error is due to the error of Q_L/Q.

3. DISCUSSION

Only the factors $S(d_{Au})$ and $h(d_{Au})$ are used for both methods in order to calculate the cross section, Therefore the comparison of the two results at 30 keV gives a measure of the accuracy. The average value is

$$\sigma_{n,\gamma}^{Au}(30\text{ keV}) = (0{\cdot}600 \pm 0{\cdot}012)\text{barn} \tag{16a}$$

$$\sigma_{n,\gamma}^{Au}(30\text{ keV}) = (0{\cdot}600 \pm 0{\cdot}009)\text{barn}. \tag{16b}$$

The result (a) contains the assumption that the difference of the two values equation (10) and equation (14) is caused by systematic errors and the result (b) that it is random. Both results contain a small correction (0·2%) to relate the cross section to the neutron energy of 30 keV, since the measurements are made with a distribution with a half-width $\Delta E \approx 10$ keV about this energy (this means an opening angle of about $2\theta \approx 18°$). Because the neutron energy spectra does not change strongly with increasing proton energy (PÖNITZ and BRUDERMÜLLER, the influence of the uncertainty of the primary energy should be small.

TABLE 3.—RESULTS OF THE RATIO $\sigma_{n,\gamma}^{Au}$ (30 keV)/$\sigma_{n,\gamma}^{Au}$ (64 keV)

Author	$\sigma_{n,\gamma}^{Au}(30\text{ keV})/\sigma_{n,\gamma}^{Au}(64\text{ keV})$
WESTON *et al.* (1961)	1·682
HARRIS *et al.* (1965)	1·680
This work	1·676
Average value	1·679

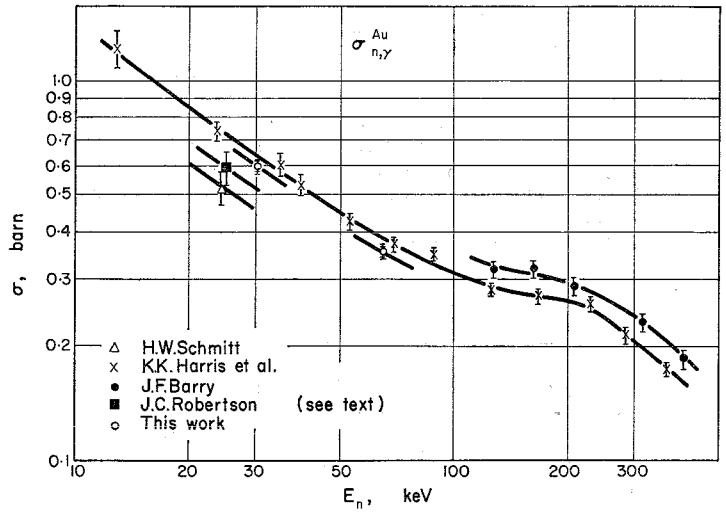

FIG. 4.—Comparison of our result with other absolute measurements.

Table 3 contains a few ratios for cross-section values at 30 and 64 keV. The average value and equation (16) lead to the result

$$\sigma_{n,\gamma}^{Au}(64\text{ keV}) = (0{\cdot}357 \pm 0{\cdot}014)\text{barn} \quad (17a)$$

$$\sigma_{n,\gamma}^{Au}(64\text{ keV}) = (0{\cdot}357 \pm 0{\cdot}009)\text{barn}. \quad (17b)$$

The meaning of equation (17a) and (17b) is the same as equation (16a) and (16b).

In Fig. 4 we compare our results with other absolute measurements in the energy region 10–300 keV. Our result is in agreement with the values measured by HARRIS *et al.* (1965) as far as the error limits overlap. The results of BARRY (1964) are about 20 per cent higher and those of SCHMITT (1963) about 25 per cent lower than the present ones. ROBERTSON (1965) has measured the (n, γ) cross section of ^{127}I at 24 keV neutron energy. His result is (0·832 $\pm$ 0·026) barn. We have used this value and the average ratio $\sigma_{n,\gamma}^{I}/\sigma_{n,\gamma}^{Au} = 1{\cdot}41 \pm 0{\cdot}10$ (GIBBONS *et al.*, 1961; KONKS *et al.*, 1964; CHAUBEY and SEHGAL 1965; SCHMITT, 1963) to determine a further $\sigma_{Au}^{n,\gamma}$ value with an accuracy < 10 per cent. This value is also included in Fig. 4.

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