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Nuclear Physics **30** (1962) 389—398, © *North-Holland Publishing Co., Amsterdam*

$Ta^{181}(p, n)W^{181}$ AND $Au^{197}(p, n)Hg^{197}$ EXCITATION FUNCTIONS BETWEEN 4 AND 13 MeV

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Received 26 September 1961

Abstract: The excitation functions for the $Ta^{181}(p, n)W^{181}$ and $Au^{197}(p, n)Hg^{197}$ reactions have been measured for incident proton energies from 4 to 13 MeV. The stacked foil method was used. The activity of the foils was measured by counting the gamma rays and X-rays emitted when the product nuclei decay by electron capture. Absolute values of the cross sections were measured at several energies for these reactions and were compared with the cross sections for compound nucleus formation given by Shapiro Threshold energies for the (p, 2n) reactions were obtained by comparing the present data with neutron yields obtained from long counter measurements. The (p, 2n) thresholds are 7 5 MeV for Ta^{181} and 8 3 MeV for Au^{197} The absolute (p, 2n) cross section values have also been obtained The total reaction cross sections above 7 MeV are compared with the optical model calculations of Bjorklund and Fernbach. The present results confirm the assumption of a surface absorption potential.

1. Introduction

The measurement of (p, n) cross sections in heavy elements in the energy region of the present work gives direct information about the proton reaction cross section since the contribution of the (p, q) charged particle cross section is negligible due to the high Coulomb barrier. The reaction cross section is determined by the imaginary part of the optical model potential. For some elements, the experimental values of the reaction cross sections make it possible to choose between a volume absorption [1]) and surface absorption [2]) potential. This is necessary because the angular distribution of elastically scattered protons or their polarizations can be obtained equally well by either one of these forms of the imaginary potential [3]).

Since there is evidence [4]) that the (p, n) interaction proceeds via the compound nucleus at these energies, the values of the (p, n) cross sections were compared with the proton capture cross sections calculated by Shapiro [5]) with $r_0 = 1.5$ fm. The total cross section for neutron production is defined as the sum of the true (p, n) cross section and the cross sections for the reactions (p, np) and (p, 2n). In the energy region of this experiment, the (p, np) cross section is negligible. Cohen [6]) and his collaborators have determined a value of about 0.5 mb for these cross sections at 13.5 MeV.

† Work performed under the auspices of the U. S. Atomic Energy Commission.

In the present work, the (p, 2n) cross section has been calculated using the compound nucleus model [7]). These values are then compared with those obtained by computing the cross section from the difference between the "long counter" [8]) and stacked foil method. Reasonably good agreement is obtained considering the relatively large experimental errors.

2. Excitation Functions

The excitation functions were measured using the well-known stacked foil technique. Stacks of 24 foils each were used in each run and for each element. Three stacks were exposed for each element at different incident proton energies using the 90-inch cm variable-energy Livermore cyclotron. The foils were 6.3 cm^2 in size and had a thickness of approximately 10 mg/cm^2. They were mounted on a brass holder and fastened to it with a Ta plate with a circular aperture of 1.9 cm diameter. The proton beam was collimated to a diameter of 1.3 cm.

The stacks were exposed for several hours to proton beams of about 0.5 μA.

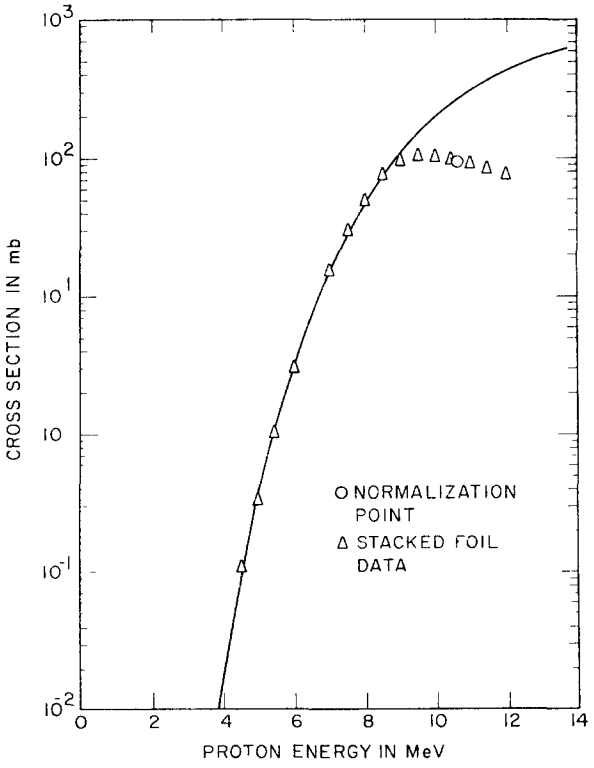

Fig 1 The $Ta^{181}(p, n)W^{181}$ cross section is shown The solid line is the cross section for compound nucleus formation using $r_0 = 1.5$ fm.

The beam energy was kept constant to about 200 keV. After the irradiations, the foils were stored to allow the short-lived activities to decay. The X-rays and gamma rays were counted by placing the foils between two NaI(Tl) scintillation spectrometers set 180° from each other. The number of counts per foil was taken as the sum of the counts given by the two crystals. This method was used to reduce errors due to a slight change in the foil position for each run. Background corrections were applied to these counting rates. The

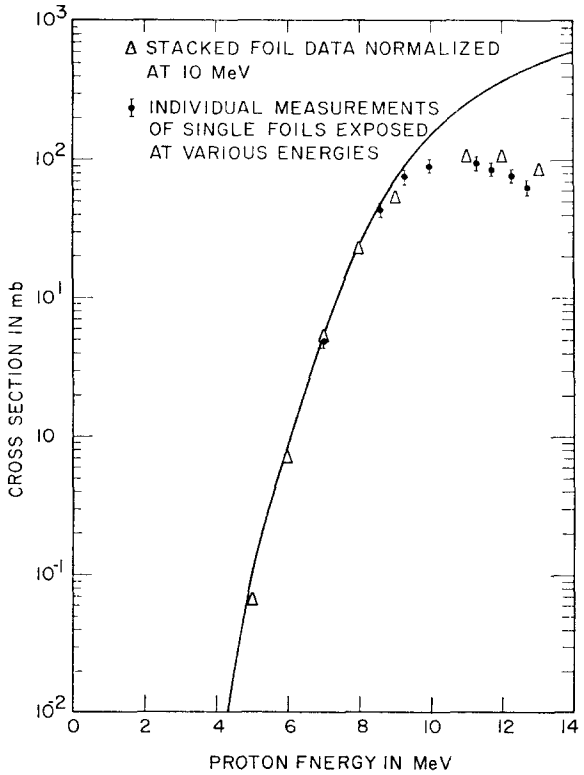

Fig. 2. The $Au^{197}(p, n)Hg^{197}$ cross section is shown. Two sets of data obtained in separate runs are plotted. The solid line is the cross section for compound nucleus formation using $r_0 = 1.5$ fm.

foils were then weighed one by one and the proton energy incident on each foil was computed using the range-energy tables [9]). The excitation functions obtained in this way are shown in figs. 1 and 2.

In the case of the $Au^{197}(p, n)Hg^{197}$ excitation function, the complex decay scheme of Hg^{197} makes it necessary to follow a slightly different procedure. This point is discussed in more detail in the following section.

3. Determination of the Cross Sections

The absolute cross sections of both reactions were measured by exposing thin gold and tantalum foils to the cyclotron beam. The energy of the beam was

chosen to correspond to regions on the excitation curves which are relatively independent of the proton enrgy so that any inaccuracies in setting the machine energy will not greatly influence the result. The beam current was measured using a carefully isolated Faraday cup and a beam current integrator circuit which was calibrated with a standard battery. The Faraday cup was protected by a guard ring at −1000 V designed to prevent secondary electrons from leaving the target and also to prevent secondaries produced in the collimator from reaching it. The number of active atoms produced on the foils by a known number of micro-coulombs of proton beam is determined by counting the induced activities of the foils. Since the decay schemes of the two isotopes are essentially different, each case is treated separately.

3.1. THE $Au^{197}(p, n)Hg^{197}$ CROSS SECTION

The $Au^{197}(p, n)Hg^{197}$ reaction [10]) populates two levels in Hg^{197}, the ground state and an isomeric level at 297 keV. The isomeric level decays to the Hg^{197} ground state through an intermediate level at 133 keV with a half-life of 24 h. No cross-over transition is observed. A small fraction (3 %) of the decays proceed directly to Au^{197}. The ground state of Hg^{197} then decays by electron capture to Au^{197} with a half-life of 65 h. The electron capture process occurs predominantly (99 %) to the 77 keV level in Au^{197}. Since both of these activities (the isomer and the ground state) are produced by the (p, n) reaction, the number of active atoms in each state must be measured. The (p, n) cross section is then computed for the population of each level and the total (p, n) cross section is the sum of the two partial cross sections.

The cross section for the production of the 24-h isomer was measured using the 133-keV gamma ray emitted following the decay of the isomer. A large (5.1 cm × 5.1 cm) NaI(Tl) crystal was used to count the gamma rays which were collimated to a diameter of 1.3 cm. The 133-keV gamma-ray line was found to decay exponentially with a mean life of 24 h so that a simple relationship exists between the counting rate and the number of active atoms at the end of the cyclotron bombardment. To obtain the correct cross section, the total conversion coefficient α_{133} of the 133 keV must be known. In the present work, the value $\alpha_{133} = 2.1$ given by Huber *et al.* [11]) was used.

The calculation of the cross section for the production of the Hg^{197} ground state is slightly more complicated. Two radiations characterize the decay of the ground state: the K X-rays emitted in the capture process and the 77-keV gamma ray from the transition of the 77-keV level in Au^{197} to the ground state. There are also L and M X-rays associated with the decay but these are not detected by the counter. The decay rate of the Hg^{197} ground state was determined by measuring the counting rate in the composite spectrum peak of the K X-rays and the 77-keV gamma rays. In order to relate this counting rate to the decay rate of the level, the fraction of K capture in the electron capture

process C_K and the internal conversion coefficient of the 77-keV level α_{77}, must be known. These numbers are obtained from published data on the decay scheme of Hg^{197}. The K capture fraction computed from the decay energy [12]) is $C_K = 0.79$. The measured internal conversion coefficient of the 77-keV level [13]) is $\alpha = 2.5$.

The cross section for the production of the Hg^{197} ground state was determined by measuring the count rate in the composite K X-ray plus the 77-keV gamma-ray peak. The 24-h isomer contributes to the K X-ray (Hg X-rays) portion of the peak, since both of the gamma rays are highly K converted. The 65-h activity contributes to the peak by K electron capture and by the emission of 77-keV gamma rays. The absolute counting rate in the peak was measured about 130 h after the end of the cyclotron exposure. At that time, the 24-h activity contributes only about 5 % of the total counting rate in the peak. Neglecting this correction for the moment, the decay rate is

$$\frac{dN(\text{K X-ray}+77\text{ keV})}{dt} = Gf\lambda_{65}\left[N_{24}(0)\frac{\lambda_{24}}{\lambda_{24}-\lambda_{65}}+N_{65}(0)\right]e^{-\lambda_{65}t}, \qquad (1)$$

where G is the geometry-efficiency factor of the system, f is a factor depending upon the K capture ratio to the 77-keV level and the internal conversion coefficient of that level, λ_{24} and λ_{65} are the respective decay constants and $N_{24}(0)$ and $N_{65}(0)$ are the number of active atoms of the 24-h isomer and the Hg^{197} ground state at the end of the cyclotron exposure. The factor containing $N_{24}(0)$ must be included because the decay of the 24-h isomer populates the Hg^{197} ground state. The value of $N_{24}(0)$ obtained from the measurement of the 133-keV gamma-ray line is used to solve eq. (1) for $N_{65}(0)$. The cross sections are then computed using the standard methods. The results are shown in table 1.

TABLE 1

$Au^{197}(p, n)Hg^{197}$ reaction cross sections

Proton energy (MeV)	σ_{24}(mb)	σ_{65}(mb)	σ_T(mb)
7.0±0.4	1.2±0 2	3 6±0.5	4.8± 0 6
8 6±0.3	12 ±2	31 ±5	43 ± 6
9.3±0 2	26 ±4	48 ±7	74 ± 8
10 0±0 2	38 ±6	52 ±8	90 ±10
11.3±0 2	53 ±8	40 ±6	93 ±10
11.7±0 2	51 ±8	34 ±5	85 ± 9
12 3±0.2	46 ±7	30 ±4	76 ± 8
12.7±0 2	40 ±6	23 ±4	63 ± 8

The cross sections for the formation of the 24-h isomer are in good agreement with the results of the Argonne group. The ground state cross sections reported here are somewhat smaller than those given by the Argonne group, however,

these numbers depend critically on the values assumed for C_K and the internal conversion coefficient of the 77-keV level. More detailed work on this isotope is required.

3 2 THE $Ta^{181}(p, n)W^{181}$ CROSS SECTION

The isotope W^{181} decays by electron capture to Ta^{181} with a half-life of 145 d. The situation in this case is considerably simpler than the one discussed in the previous section. The spectrum observed from this source is given in ref. [14]) and is quite simple, containing only the L and K X-rays. The isotope does decay to two excited levels in Ta^{181} at 137 keV and 152 keV but only 0.3 % of the decays proceed in this manner. The L to K ratio has been measured [14]) and since no other lines appear the determination of the cross section is simple. The cross section at an incident proton energy of 10.6 MeV is

$$\sigma = 96 \pm 10 \text{ mb}.$$

The excitation function has been normalized at 10.6 MeV using this value of the cross section.

4. Discussion of Results

Figs. 1 and 2 show the experimental (p, n) cross sections obtained in the present work compared with the proton cross sections for the formation of the compound nucleus with $r_0 = 1.5$ fm as calculated by Shapiro [5]). The (p, n) cross section measured by activation gives what sometimes is called the "true" σ(p, n) corresponding to the process A+p → B+n. The agreement between the true σ(p, n) and the calculated proton capture cross sections at low energies confirms the experimental fact that no other reactions are taking place. Around 8 MeV the cross sections begin to diverge. The difference between the two curves grows larger as the energy increases. Since the proton-charged particle cross section σ(p, q) is still quite small at 14 MeV, the proton cross section for the formation of the compound nucleus corresponds to the total (p, n) cross section

$$\sigma(\text{p, n})_{\text{total}} = \sigma(\text{p, n})_{\text{true}} + \sum_i \sigma(\text{p, n}, i). \tag{2}$$

The second term on the right-hand side represents all other neutron-producing cross sections. The (p, np) process is negligible [6]) at these energies, and the only important contribution therefore comes from the (p, 2n) cross section.

To evaluate the (p, 2n) cross section theoretically, the threshold energy for this reaction must be known. An experimental value for this threshold was obtained by comparing the (p, n) cross section measured by activation and the neutron production cross section measured with a long counter [8]) for these

nuclei. The long counter cross section is given by

$$\sigma(p, n)_{LC} = \sigma(p, n)_{true} + 2\sigma(p, 2n). \tag{3}$$

The slopes of the curves obtained using activation and long counter methods will be identical up to an energy corresponding to the (p, 2n) threshold. Above this energy the difference between the long counter neutron production cross section and the activation cross section is twice the (p, 2n) cross section.

Following this procedure, a threshold of 7.5 MeV was found for $Ta^{181}(p, 2n)W^{180}$ and 8.3 MeV for $Au^{197}(p, 2n)Hg^{196}$. The (p, 2n) cross sections obtained experimentally in this way were compared with the predictions of the statistical model of the compound nuclei. The (p, 2n) cross section is given by

$$\sigma(p, 2n) = \sigma(p, n)_{total}\left[1 - \left(1 + \frac{\varepsilon_c}{\Theta}\right)\exp\left(-\frac{\varepsilon_c}{\Theta}\right)\right], \tag{4}$$

where ε_c is the excess energy over the threshold of the (p, 2n) reaction and Θ is the nuclear temperature of the intermediate nucleus B in the process

$$p + A \rightarrow B + n \rightarrow C + n + n. \tag{5}$$

The temperature Θ is given by $\Theta = (E/a)^{\frac{1}{2}}$ if the nuclear level density is given by $\omega(E) = Ce^{2\sqrt{aE}}$, for a Fermi gas model of the nucleus. Here C and a are constants and E is the maximum excitation energy of intermediate nucleus B, given by

$$E = T_p + Q - \delta. \tag{6}$$

The quantity T_p is the kinetic energy of the incident proton, Q the excess energy of the reaction A(p, n)B and $\delta = \delta_Z + \delta_N$ the pairing energy of the nucleons in B. The pairing energies were taken from Cameron [15]). The density parameter used in the calculations was obtained experimentally from the slope of the line $\ln[N(\varepsilon_n)/\delta(\varepsilon_n)\varepsilon_n]$ as a function of $\sqrt{E}$ using the data of Anderson *et al.* [16]).

Table 2 shows the values of the different parameters used in these calculations. The (p, n) total cross sections were taken equal to the proton capture cross section given by Shapiro [5]).

TABLE 2

Parameters used in the (p, 2n) cross-section computation

Isotope	r_0(fm)	a	E_{th}(p, 2n) (MeV)	Q(p, n) (MeV)	δ(MeV)
Ta^{181}	1 5	181/22 5	7 5	−0 260	1 23
Au^{197}	1 5	197/22.5	8.3	−0.750	0 72

The level density can also be represented [7]) by the function $Ce^{-\varepsilon_n/\Theta}$. This expression implies that the nuclear temperature Θ is a constant independent

of the excitation energy. In this case Θ can be obtained experimentally from the slope of the line $\ln[N(\varepsilon_n)/\sigma_n(\varepsilon)\varepsilon_n]$ as a function of ε_n. The value of Θ measured[16]) in this way was 0.75 MeV.

Table 3 gives the calculated (p, 2n) cross sections according to expression (4) for both level density functions and the experimental values obtained

TABLE 3

Comparison of experimental and calculated (p, 2n) cross sections

Proton energy (MeV)	$Ta^{181}(p, 2n)W^{180}$ cross sections (mb)			$Au^{197}(p, 2n)Hg^{197}$ cross sections (mb)		
	Calculated		Experimental	Calculated		Experimental
	a = const	T = const.		a = const	T = const	
8	5.13	6 81	3.4± 0 5			
9	50 5	65 4	40 ± 7	12.3	16.8	23± 4
10	139	169	148 ±27	77 7	99.3	73±13
11	261	298	404 ±54	183	219	150±27
12	388	327	460 ±83			

from the half-difference between the long counter (p, n) cross sections minus the activation (p, n) cross section. Both level density functions agree with the neutron spectrum [16]) from the (p, n) reactions in Ta and Au. However, the (p, 2n) cross sections calculated with a constant nuclear temperature are consistently higher than those calculated with a constant nuclear density parameter, which seem to give better agreement with the experimentally measured cross sections. This better agreement will favour the level density given by $\omega(E) = C \exp[2(aE)]^{\frac{1}{2}}$.

The reaction cross sections for these two nuclei, expressed as the sum of the (p, n) cross section measured by activation plus the (p, 2n) cross section calculated from the compound nucleus model were compared with the theoretical calculations of Bjorklund and Fernbach † for the reaction cross section given by the optical model using a surface absorption potential [2]).

Figs. 3 and 4 show the comparison between experiment and theory. The parameters used in the optical model calculations are the following:

$$r_0 = 1.25 \text{ fm}, \qquad a = 0.65 \text{ fm}, \qquad b = 1.2 \text{ fm}, \qquad V = f_1(T_p) + Z/A^{\frac{1}{3}} \text{ MeV},$$
$$W = f_2(T_p) \text{ MeV},$$

where $f_1(T_p)$ and $f_2(T_p)$ are two functions of the incident proton energy given by Bjorklund and Fernbach [2]). The spin orbit potential is 20 times the Thomas term. All these parameters have been obtained from elastic scattering and polarization data.

† Private communication.

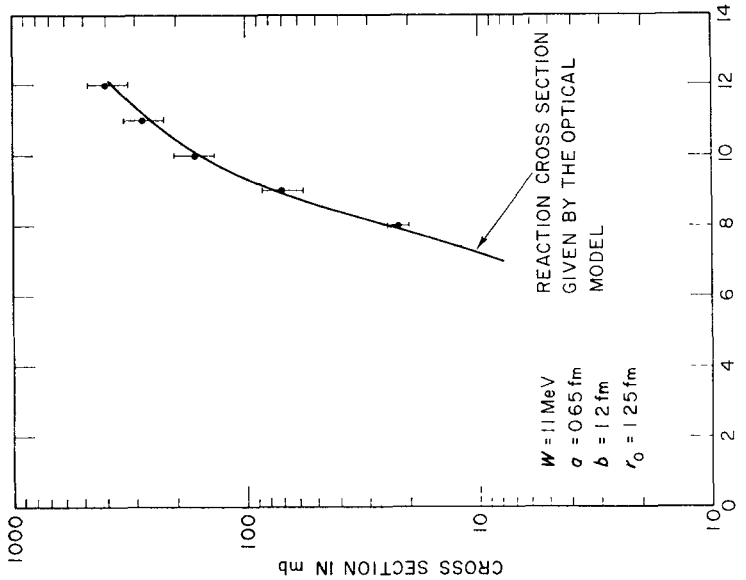

Fig. 4. The total reaction cross section for Au^{197}, similar to that of fig. 3, is shown

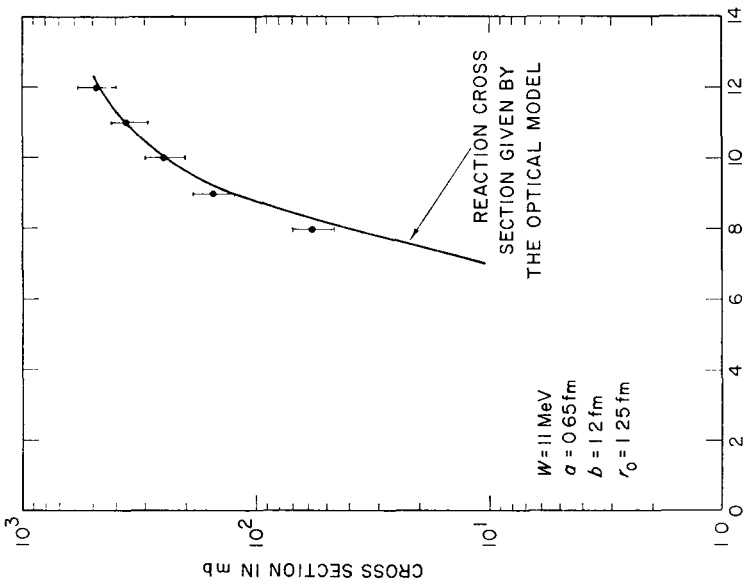

Fig. 3. The total reaction cross section $\sigma_R = \sigma(p,n) + \sigma(p,2n)$, computed using the optical model, is shown for Ta^{181}. The method of obtaining experimental points is described in the text

The good agreement between theory and experiment as shown in figs. 1 and 2 and in table 3 indicates that, in heavy nuclei at the energies of this experiment, the compound nucleus model adequately explains the reaction mechanism. Furthermore, the agreement between the reaction cross sections shown in figs. 3 and 4 confirms the choice of a surface absorption potential in the optical model of the nucleus as the correct one. The reaction cross sections predicted by the optical model using a volume potential are much too small. For example, at 10 MeV, the total reaction cross sections for Ta and Au, using a volume absorption potential, are about 150 mb and 80 mb, respectively, compared to 225 mb and 150 mb obtained experimentally. Similar conclusions favouring a surface absorption potential have been reached by Nodvick *et al.*[17]) and Albert[8]) for other nuclei.

The authors would like to thank Messrs. LeRoy Erickson, D. R. Rawles and the crew of the 90-inch Cyclotron for their cooperation during this experiment.

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