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ASSOCIATION EURATOM - C.N.E.N.
(European Atomic Energy Community and Comitato Nazionale
per l'Energia Nucleare)

CROSS-SECTION MEASUREMENT OF (n, 2n) REACTIONS ON 14,1 MeV NEUTRONS

by

M. CEVOLANI and S. PETRALIA
(Istituto Nazionale di Fisica Nucleare - Sezione di Bologna)

1963

Work performed by Istituto Nazionale di Fisica Nucleare -
Sezione di Bologna
under the Euratom contract No. 001-60-12 MPAI

Reprinted from
IL NUOVO CIMENTO
Serie X, Vol. 26 - 1962

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M. CEVOLANI - S. PETRALIA
16 Dicembre 1962
Il Nuovo Cimento
Serie X, Vol. **26**, pag. 1328-1338

Cross-Section Measurement of (n, 2n) Reactions on 14.1 MeV Neutrons.

M. CEVOLANI and S. PETRALIA

Istituto Nazionale di Fisica Nucleare - Sezione di Bologna

(ricevuto il 26 Luglio 1962)

Summary. — We have measured the absolute cross-section of the reaction (n, 2n) in twelve nuclei having β^+ residual activity, using neutrons of 14.1 MeV. The results are compared with data existing in the literature and are discussed with reference to the theory of Weisskopf and Ewing.

1. – Introduction.

The measurement of the cross-section of reaction (n, 2n) in atomic nuclei, with neutrons of energy around 14 MeV has been the object of many papers recently appeared. It is generally noted that results are often disagreeing, beyond the limits of error quoted by the single authors. This happens as well for low mass number elements where the cross-section is generally small, as for medium and heavy elements for which there are cross-sections of the order of 1 barn.

Measurements have been made principally with the activation method. The activity of the neutron-irradiated sample has been determined either counting directly the β-rays emitted by the residual nucleus in a given time interval, or counting the γ-rays, or, also, counting γ-ray annihilation coincidences in case of nuclei showing β^+ activity.

There are scanty data on the variation of this cross-section with the energy of neutrons; the determination has mostly been made for one single energy only, different for the different authors.

The method of measurement with all the corrections to do and errors to estimate as well as the quick variation of the cross-section near the reaction threshold, where till now measurements were nearly always taken, may justify the divergence of experimental data. In experiments with the activation method, the value of the cross-section may be influenced by the incertitude on the decay scheme and on the half-life of the residual nucleus.

In view of the interest which the knowledge of cross-sections of reactions (n, 2n), may offer for reactor technique as well as the disagreement still existing in experimental values of these cross-sections and the importance of the knowledge of their exact values for the comparison of theories on nuclear reactions we have undertaken a series of measurements of these cross-sections which we report in the following.

2. – Experimental technique.

Neutrons were produced bombarding a target of tritium adsorbed in zirconium, with a deuteron beam accelerated by a potential difference of 240 kV. In order to avoid a strong energy variation with emission angle and deuteron energy of the neutrons impinging on the sample, this was set at an average angle of 90° to the direction of the deuteron beam, and it was kept small (a little cylinder of $(7 \div 8)$ mm diameter). In case of solid substances the thickness of the cylinder could vary between 1 and 8 mm; in case of powders, we adopted a cylindrical plexiglas container with very thin walls, and a length of 8 mm. The average distance of the sample from the tritium target was 5 cm. The utilized neutron energy was therefore (14.13 ± 0.1) MeV.

The neutron flux was monitored by a plastic scintillator, whose indications served to regulate the intensity of the deuteron current on the tritium target, in order to keep the neutron flux constant throughout the irradiation. Irradiation lasted generally three or four half-lifes of the residual nucleus, but never more than 5 hours. The value of the neutron flux necessary to determine the cross-section was determined counting the α-particles associated with the neutron-producing reaction through a solid state detector, 1.3 m distant from the tritium target so as to take up the α-particles emitted at 90° to the direction of the deuteron beam. In Fig. 1 is shown the spectrum of these particles taken with the 200 channel analyser.

The activity of residual nuclei was determined counting the γ-γ annihilation coincidences. We have therefore limited our study to nuclei with β^+ residual activity. The sample was introduced, within a minute from irradiation, in a brass cylinder whose walls of appropriate thickness served to the total conversion of the β^+. This cylinder was centered along the axis of two NaI(Tl) crystals 2 in. high and 2 in. in diameter situated at 8 cm distance. The electric pulses of the photomultipliers connected with the crystals were sent to a fast-slow coincidence apparatus; the differential discriminators of the slow channels were so regulated as to let pass the photoelectric peak corresponding to the radiation of 0.511 MeV; the position of this peak was continuously controlled with the 200 channel analyser.

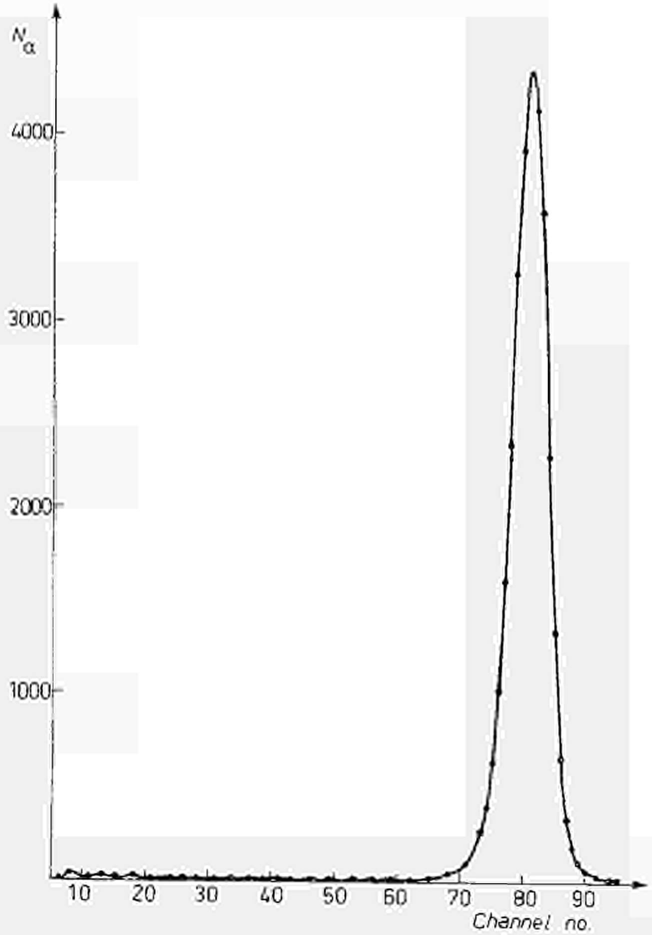

Fig. 1. – Pulse height distribution of α-particles from the reaction T(D,n)α.

The calibration of the apparatus was done with the annihilation γ-rays from a ^{22}Na source, spread on paper in a little plexiglas cylinder of the identical dimensions of those bearing the irradiated samples. These sources had been calibrated by us after the absolute method of Meyer, Schmid and Huber (¹). The counter's efficiency was corrected for the effect of the spurious coinci-

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(¹) K. P. Meyer, P. Schmid and P. Huber: *Helv. Phys. Acta*, **32**, 425 (1959) dences between photoelectrons of the annihilation radiation and Compton electrons of the 1.276 MeV radiation emitted in cascade with the β^+ of ^{22}Na. For this correction we assumed that the detection probability of the Compton spectrum of the 1.276 MeV radiation, which corresponds to the photoelectric peak of the 0.511 MeV radiation, remains almost unaltered for slightly stronger energies; it is then easy to obtain the Compton contribution inside the discriminator channels.

The number of coincidences, experimentally determined for each sample, was corrected for the loss of coincidences due to absorption of annihilation radiation in the sample. Therefore, a ^{22}Na source in form of a thin film, was counted in correspondence of the photoelectric peak of 0.511 MeV, first alone inside the converter, and then together with the nonactivated samples: the intensity reduction so obtained was taken as the correction to apply to the registered coincidences with the activated sample.

Possible error sources in our measurements are the following:

1) Absolute measurement of the neutron flux by the detection of the recoil α-particles; the corresponding error is estimated to be not over 4%, taking account also of eventual scattering of neutrons on the sample from the material surrounding the tritium target.

2) Uncertainty in the sample's geometry in respect of the neutron source; this contributes an error of about 3%.

3) The error in the calibration of the detector with ^{22}Na sources is also estimated to be 3%.

4) The error in counting γ-γ coincidences from the activated sample is to be referred to the uncertainty of the sample's position (about 2%) and to statistical error. This last one varies from element to element and may go from 1% to 6%.

5) Error in the number of irradiated nuclei owing to impurities, errors in weighing, or uncertainty in the isotopic concentration: this error is less than 1%.

Decay schemes and decay constants for the activated nuclei were taken from the tables of KUNZ and SCHINTLMEISTER (2). Many of the decay constants have been controlled by us. We did not attribute errors to the tabulated values and have neglected errors in the measurement of irradiation times and of the time elapsed between end of irradiation and begin of count.

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(2) K. KUNZ and J. SCHINTLMEISTER: *Tabellen der Atomkerne* (Berlin, 1959).

3. – Results.

The cross-section has been calculated with the well-known formula

$$(1) \qquad \sigma = \frac{h t_1}{n b N_\alpha} \, \frac{\exp[\lambda t_2]}{(1-\exp[-\lambda t_1])} \, \frac{\lambda C_{\Delta t}}{(1-\exp[-\lambda \Delta t])} \, R \, ,$$

where t_1 is the time of neutron-irradiation of the sample; N_α the number of α-particles counted in this time; h a constant of the apparatus which allows to pass from the number of α-particles to the neutron flux in the median plane of the sample; n the number of nuclei of the chosen isotope per cm² of sample; b the β^+ fraction of the activity of the residual nucleus; λ the decay constant of the nucleus; t_2 the time elapsing between the end of irradiation and the beginning of the activity measurement; $C_{\Delta t}$ the number of coincidences registered in time Δt and R the calibration constant of the annihilation-radiation detector, determined with the use of the calibrated ^{22}Na sources.

The results of the measurements are shown in Table I. Then are listed the chemical compounds containing the examinated nuclides, the half-lifes of

Table I. – *Measured* (n, 2n) *cross-section.*

Nuclid	Chemical	Half-life	σ(n,2n) (mb)	Exp. errors ($\pm$mb)	Measurements by other authors (mb)
^{14}N	melanine	10,05 min	5,4	0.46	7.41 (3); 5.18 (4)
^{19}F	teflon	112 min	38.9	2.3	51.9 (3)
^{31}P	element	2.53 min	5.1	0.45	10.9 (3); 8.7 (4)
^{45}Sc	Sc_2O_3	3.92 h	130	7.8	150 (5); 198 (3); 148 (6)
^{46}Ti	element	3.08 h	13.3	1.1	13 (5); 31.8 (3)
^{63}Cu	element	10.1 min	409	25	see Table II
^{64}Zn	element	38.3 min	105	7	119 (7); 167 (3); 107 (8)
^{69}Ga	Ga_2O_3	67 min	735	44	923 (3); 1070 (6)
^{79}Br	CBr_4	6.4 min	793	48	835 (3)
^{92}Mo	element	15.7 min	106	7.5	211 (3); 315 (9); 132 (7)
^{107}Ag	element	24 min	734	44	889 (3); 657 (6); 458 (7)
^{141}Pr	Pr_6O_{11}	3.4 min	1240	74	1801 (3); 1378 (6); 1386 (4)

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(3) L. A. Rayburn: *Phys. Rev.*, **122**, 168 (1961).

(4) J. M. Ferguson and W. E. Thompson: *Phys. Rev.*, **118**, 228 (1960).

(5) R. J. Prestwood and B. P. Bayhurst: *Phys. Rev.*, **121**, 1438 (1961).

(6) C. S. Khurana and H. S. Hans: *Nucl. Phys.*, **28**, 560 (1961).

(7) S. Yasumi: *Journ. Phys. Soc. Japan*, **12**, 443 (1957).

(8) E. Weigold and R. N. Glover: *Nucl. Phys.*, **32**, 106 (1962).

(9) P. Strohal, N. Cindro and B. Eman: *Nucl. Phys.*, **30**, 49 (1962).

residual nuclei, the values of the cross-sections and their errors. The last column shows the most recent values for σ to be found in the literature, possibly chosen for a neutron energy of 14.1 MeV.

The values obtained by us result, generally, to be somewhat lower than those reported by other authors. Comparison is difficult as the measurements are made at different neutron energies and with different techniques. We shall limit therefore our attention to the data of FERGUSON and THOMPSON (4), RAYBURN (3), GLOVER and WEIGOLD (8,10), who have used the method of the annihilation γ-coincidences and have made measurements at different energies, and further to the measurements of PRESTWOOD and BAYHURST (5) who have made for many nuclei σ-measurements as function of the neutron's energy. It is interesting to note that for the nuclei: ^{14}N, ^{31}P, ^{63}Cu, ^{141}Pr our σ-values are different by less than 18% from those of FERGUSON and THOMPSON; for ^{45}Sc, which goes to the fundamental state ^{44}Sc (Fig. 2), and for ^{46}Ti the agreement of our values with those of PRESTWOOD and BAYHURST is still better. For ^{64}Zn our σ of 105 mb agrees very well with the value 107 mb given by GLOVER and WEIGOLD at the same neutron energy. The σ(n, 2n) which we have measured for ^{107}Ag falls between those measured by RAYBURN and by

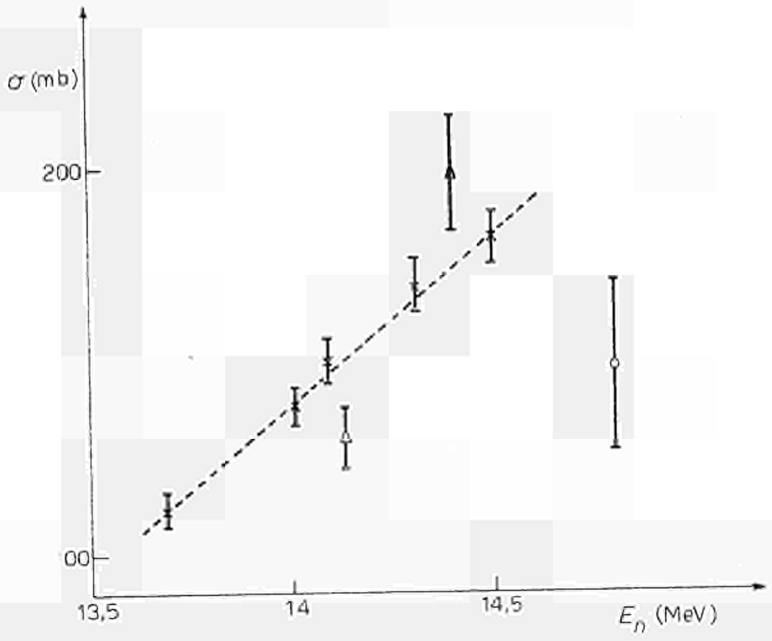

Fig. 2. – Experimental cross-section for ^{45}Sc(n, 2n) reaction *vs.* neutron energy. × PRESTWOOD and BAYHURST; ▲ RAYBURN; o KHURANA *et al.*; △ present work.

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(10) R. N. GLOVER and E. WEIGOLD: *Nucl. Phys.*, **29**, 309 (1962).

Khurana and **Hans** (⁶) at neutron energies of 14.4 and respectively 14.8 MeV. The situation in the case of ^{92}Mo is shown in Fig. 3, where in correspondence with the neutron energy at which the measurement was made are reported the σ-values obtained by us, by **Rayburn**, by **Yashumi** (⁷) and by **Strohal**, **Cindro** and **Eman** (⁹).

Follwoing **Brolley**, **Fowler** and **Schlacks** (¹¹) it seems that through the reaction (n, 2n) the isomeric state of ^{91}Mo is not activated; also some tentatives we made to find this state remained unfruitful.

The σ(n, 2n) for ^{63}Cu has often been used as reference cross-section and its

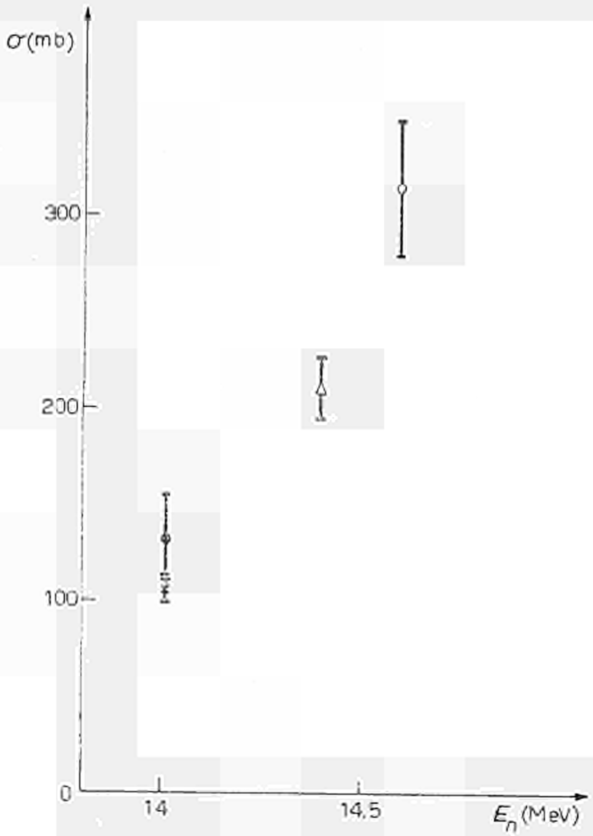

Fig. 3. – Experimental cross-section for ^{92}Mo(n, 2n) reaction *vs.* neutron energy. ∆ Rayburn; ● Yasumi; o Strohal *et al.*; × present work.

Table II. – ^{63}Cu σ(n, 2n) ^{62}Cu (mb).

Authors					
E_n	Ferguson and Thompson	Yasumi	Glover and Weigold	Present work	Rayburn (value adopted)
13.77 ± 0.2	378 ± 34	—	—	—	—
13.86 ± 0.1	—	—	424 ± 21	—	—
14.11 ± 0.1	—	—	455 ± 23	—	—
14.13 ± 0.1	—	—	—	409 ± 25	—
14.1	—	556 ± 28	—	—	—
14.37 ± 0.15	—	—	488 ± 24	—	—
14.4 ± 0.3	—	—	—	—	503 ± 57

(¹¹) J. E. **Brolley**, J. L. **Fowler** and K. L. **Schlacks**: *Phys. Rev.*, 88, 618 (1952).

knowledge has therefore a special importance. In Table II are reported its values in mb at different energies around 14 MeV.

4. – Discussion.

The cross-section for the reaction (n, 2n) has been calculated by WEISSKOPF and EWING (¹²) basing on the compound nucleus model. It is given by

$$\sigma(\text{n},2\text{n})=\sigma(\text{n},\text{n}')\left[-(1+\varepsilon/T)\exp\left[-\varepsilon/T\right]\right], \tag{2}$$

where $\sigma(\text{n},\text{n}')$ is the cross-section for the emission of at least one neutron from the compound nucleus, or the cross-section for inelastic scattering; T is the nucleus' temperature resulting from the emission of the first neutron (here the target nucleus), ε the difference between the energy of the incident neutrons and the threshold energy of the reaction (n, 2n). The cross-section $\sigma(\text{n},\text{n}')$ is often substituted by $\sigma_t(\text{n})$, cross-section for the formation of the compound nucleus by neutron collision.

Owing to the absence of exact data on $\sigma(\text{n},\text{n}')$ and on T, it is not easy to calculate $\sigma(\text{n},2\text{n})$ using eq. (2). One may obtain $\sigma(\text{n},\text{n}')$ from the experimental values of the nonelastic cross-section σ_{ne}, subtracting from it the cross-sections $\sigma(\text{n},\text{p})$ and $\sigma(\text{n},\alpha)$ for the emission of protons and α-particles respectively, which however are not always known. Here we have utilized the σ_{ne} measured by FLEROV and TALYZIN (¹³) for neutrons of energy $=14.5$ MeV, and in the absence of experimental data, we have calculated σ_{ne} with the expression of Flerov and Talyzin:

$$\sqrt{\frac{\sigma_{\text{ne}}}{\pi}}=(1.2A^{\frac{1}{3}}+2.1)\cdot 10^{-13}\ \text{cm}, \tag{3}$$

which agrees well with experience. We have taken $\sigma(\text{n},\text{p})$ from the recent paper of GARDNER (¹⁴) and the $\sigma(\text{n},\alpha)$'s for light elements from the literature, while for the heavy elements we have neglected its value. For ^{46}Ti, as we held the only existing experimental value of $\sigma(\text{n},\text{p})$ not to be reliable we have used $\sigma_c(\text{n})$ calculated after BLATT and WEISSKOPF (¹⁵).

The temperatures of the target nuclei were calculated with the relation

$$T=(E_{\text{n}}/a)^{\frac{1}{2}}, \tag{44}$$

(¹²) V. F. WEISSKOPF and D. H. EWING: *Phys. Rev.*, **57**, 472 (1940).

(¹³) N. N. FLEROV and V. M. TALYZIN: *Journ. Nucl. Energy*, **4**, 529 (1957).

(¹⁴) D. G. GARDNER: *Nucl. Phys.*, **29**, 373 (1962).

(¹⁵) J. M. BLATT and V. F. WEISSKOPF: *Theoretical Nuclear Physics* (New York, 1952).

with E_n the energy of the impinging neutrons and a the density parameter of the nuclear levels. This parameter has been calculated on the basis of many nuclear models (16). In Table III are reported, besides the values of $\sigma(n, n')$ and of the threshold energies of the reaction, the values of $\sigma(n, 2n)$ calculated taking $a = A/13$ and $a = A/22$.

TABLE – III. *Calculated* (n, 2n) *cross-sections* (barn).

Nuclide	$\sigma(n, n')$	E_{th} (threshold-energy) (MeV)	$\sigma(n, 2n)$ $a = A/13$	$\sigma(n, 2n)$ $a = A/22$
^{14}N	0.62	11.30	0.113	0.075
^{19}F	0.78	10.80	0.22	0.15
^{31}P	0.92	12.78	0.095	0.061
^{45}Sc	1.22	11.57	0.43	0.31
^{46}Ti	1.29 (*)	13.48	0.051	0.031
^{63}Cu	1.35	10.82	0.77	0.59
^{64}Zn	1.28	11.90	0.48	0.34
^{69}Ga	1.51	10.30	1.02	0.80
^{79}Br	1.56	10.77	1.004	0.78
^{92}Mo	1.74	12.48	0.55	0.38
^{107}Ag	1.84	9.10	1.65	1.46
^{141}Pr	2.06	9.70	1.85	1.64

(*) $\sigma_c(n)$.

From ^{45}Sc, $\sigma(n, 2n)$ calculated with $a = A/22$ agree well with the experimental data. For ^{45}Sc the calculation gives the total cross-section, sum of the cross-sections which lead to ^{44}Sc in the isomeric as well as in the fundamental state. The total cross-section at $E_n = 14.09$ MeV results, following PRESTWOOD and BAYHURST, to be 266.2 mb. The cross-sections calculated for ^{64}Zn and ^{92}Mo are greater than the experimental ones by a factor 3. This latter nucleus is a magic one in the number of neutrons ($N = 50$). This may be due to the fact that the compound nucleus decays, following BLATT and WEISSKOPF, before thermal equilibrium is reached; only few nucleons would then receive energy from the impinging neutron and the emission of one neutron with relatively high energy would leave the nucleus in a low-excitation state. In other words it would be a kind of direct interaction. The reaction mechanism with direct interaction has been invoked by FERGUSON and THOMPSON to justify the disagreement between experimental results and eq. (2), in analogy with the most recent opinions on the reactions with emission of charged particles.

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(16) I. DOSTROVSKI, P. RABINOWITZ and R. BIVINS: *Phys. Rev.*, **111**, 1659 (1958).

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For nuclei with low mass number the disagreement between theory and experiment is certainly sharper. Here, however, cross-sections for emission of charged particles are very large. **Weigold** and **Glover** have suggested an applicability criterium for eq. (2) based on the ratio of the emission probabilities of a proton and a neutron from the compound nucleus. Equation (2) would be valid if this ratio resulted to be less than 0.01.

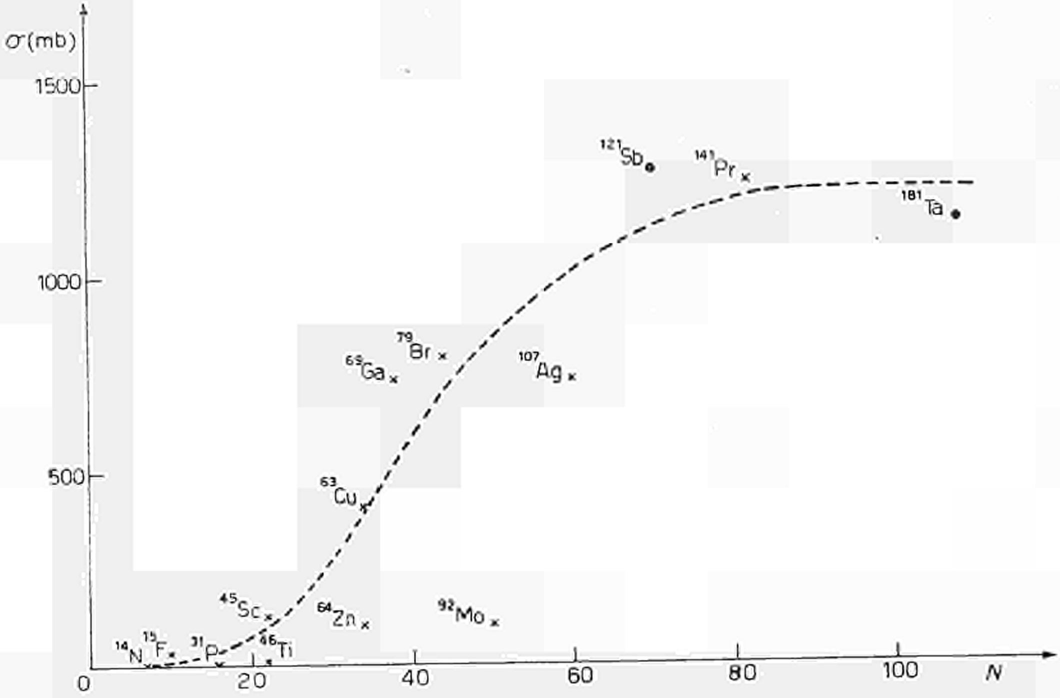

Fig. 4. – Measured values of (n, 2n) cross-sections plotted against neutron number N
× present work; ● **Prestwood** and **Bayhurst**.

In Fig. 4 we have ordered the σ(n, 2n) measured by us as functions of the number of neutrons contained in the nuclei. We have added also the cross-sections for ^{121}Sb and ^{181}Ta taken from **Prestwood** and **Bayhurst**. One notes a rather fast increase of σ between scandium ($N = 24$) and gallium ($N = 38$) followed by a slower increase towards praseodymium. In this region an anomaly is presented by the σ referring to the magic nucleus ^{92}Mo, whereas ^{141}Pr, which is also magic ($N = 82$), seems to behave normally.

We note finally that in the three pairs of nuclides ^{45}Sc and ^{46}Ti, ^{63}Cu and ^{64}Zn, ^{90}Zr and ^{92}Mo, whose components have the same number of neutrons, the nuclei with higher mass number have the lesser cross-section. The value for ^{90}Zr is to be found in literature (^{3,5,9}).

* * *

This work has been done on the contract Euratom/CNEN/INFN. The authors wish to thank Prof. P. Bassi, Director of the Section of Bologna, for the untiring interest which he has shown in following our toil. Messrs. G. Baldini and G. C. Degli Esposti of INFN have given constant technical assistance and we wish to express also them our deep gratitude.

RIASSUNTO

Abbiamo misurato la sezione d'urto assoluta della reazione (n, 2n) in dodici nuclei che hanno attività residua β^+, usando neutroni di 14.1 MeV. I risultati sono confrontati con i dati esistenti nella letteratura e sono discussi secondo la teoria di Weisskopf ed Ewing.

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16 Dicembre 1962
Il Nuovo Cimento
Serie X, Vol. **26**, pag. 1328-1338