THE
PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 75, NO. 12 JUNE 15, 1949

Capture Cross Sections for Fast Neutrons

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(Received January 27, 1949)

The neutron capture cross sections at an effective energy of one Mev have been measured for 32 isotopes, including 4 isomeric pairs. Unmoderated fission neutrons were used as the source, and the high intensities available by this method have made it possible to measure fast cross sections as low as 10^{-4} barn. The cross sections (except those for a few "magic number" nuclei) show a smooth variation with atomic weight, ranging from 10^{-1} b at $A=100$ to 10^{-4} b at $A=20$. The rapid change in cross section is a measure of the change in level spacing of the compound nucleus formed after neutron capture. A survey is made of all the existing data on absorption cross sections as functions of A and neutron energy.

I. INTRODUCTION

CROSS-SECTION measurements made with neutrons from chain-reacting piles have been mainly those at or near thermal energy. Although piles have a high flux of neutrons extending all the way from fission energies to thermal the great spread in energy has made cross-section measurements at specific energies exceedingly difficult. It was the purpose of the present measurements to isolate a particular group of fast neutrons, the nascent fission neutrons of energy about one Mev, and use them for measurement of absorption cross sections. Although the fission neutrons themselves show a large spread in energy, the measured cross sections can be correctly interpreted because of the smooth variation of cross section with energy in the one-Mev region. The absorption cross section for thermal neutrons varies irregularly from isotope to isotope depending on the proximity of the closest resonance level. As the neutron energy is raised, however, the width and density of the levels increase until even with a monoenergetic neutron source the observed cross section becomes an average over several levels. The absorption cross section at such high energies thus depends on the average behavior of the resonances, and is expected to be a slowly varying function of neutron energy and atomic weight. The disadvantage of the energy spread is further compensated by the large available flux, absolute calibration, and steadiness of the pile as a neutron source.

The theory of resonance absorption of neutrons in terms of the "Breit-Wigner" formula has been treated by many authors. The discussion of Feshbach, Peaslee, and Weisskopf[1] is quite convenient for the present measurements. FPW show that the average absorption cross section for fast neutrons in a small energy band ΔE at E is given by

$$\sigma_a{}^l=\frac{\pi}{k^2}(2l+1)\frac{2\pi(\Gamma_{nl}\Gamma_{al})_{\rm av}}{D_l[(\Gamma_{nl})_{\rm av}+(\Gamma_{al})_{\rm av}]} \qquad (1)$$

where $\sigma_a{}^l$ is the contribution to the cross section of neutrons with angular momentum lh and wave number k. Γ_{nl} and Γ_{al} are the neutron and absorption widths for the many levels involved ("av" signifies average value) and D_l is the level spacing. Considering only $l=0$, it is seen that where $\Gamma_n>\Gamma_a$ (neutron energy >10 kev) the variation of cross section with energy will be as $1/E$. As the neutron energy is raised, the capture cross section falls off less rapidly as higher l scattering becomes important. The onset of inelastic scattering, however, decreases the capture cross section and the energy dependence at one Mev will be the resultant of the higher l and the inelastic scattering effects. Any variation of cross section from isotope to isotope in

[1] H. Feshbach, D. C. Peaslee, and V. F. Weisskopf, Phys. Rev. **71**, 145 (1947).

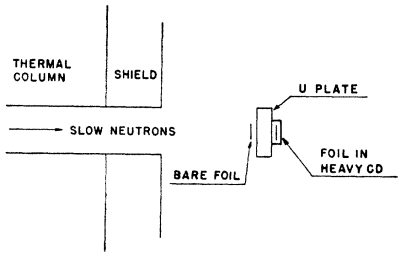

FIG. 1. Apparatus for measurement of activation cross sections for fission neutrons.

this energy region would reflect the variation mainly in level spacing, D. Thus it would be expected that the cross section at one Mev would increase with atomic weight rapidly at first (as D decreases with increasing complexity of the nucleus) then would change only slowly for heavy nuclei (as D changes slowly—see FPW, p. 155). Of course any unusual values of the neutron binding energy might be expected to give unusual values of D and hence of cross section. Measurements of absorption cross sections thus give information on the average level behavior, particularly the level spacing, D. It is because of this dependence of the cross section on the average level behavior that the spread in energy of the fission neutron distribution does not prove to be a serious complicating factor in the present measurements.

At the time the present experiments were begun, some experimental results were available on absorption cross sections for neutrons in the region of one Mev, based on work with cyclotrons, Van de Graaffs, Ra-α-Be, and photo-neutron sources. The reported cross sections were mainly for heavy nuclei (large cross sections) and in general were quite uncertain in regard to absolute calibration because of the difficulty of measuring fast neutron flux, although some of the Los Alamos results were based on careful flux calibration. The present method was devised to improve the absolute cross-section values and to extend them to the light elements for which no values existed.

TABLE I. Neutron cross sections at 1 Mev effective energy.

Isotope	Activity	Isotopic Activation Cross-Sections Thermal*	Fast	Remarks
Na^{23}	14.8 hr.	0.55 barns	0.29 mb	
Mg^{26}	10.2 min.	0.049	0.60	
Al^{27}	2.4 min.	0.23	0.40	
Cl^{37}	37 min.	0.61	0.80	
K^{41}	12.4 hr.	1.0	2.9	
V^{51}	3.9 min.	5.5	2.2	
Co^{59}	10.7 min.	0.72	0.23 } 9.2	The total σ_a is based on the assumption that the isomeric ratio of 30/0.72 holds for fast neutrons.
	5 yr.	30	— }	
Mn^{55}	2.6 hr.	11.5	3.5	
Ni^{64}	2.6 hr.	3.4	6.7	See text
Cu^{63}	12.8 hr.	3.1	8.9	
Cu^{65}	5 min.	2.0	6.0	
Zn^{68}	57 min.	0.89	7.1 } 20.1	Isomeric ratio: Thermal 10.5; fast 0.55
	13.8 hr.	0.085	13.0 }	
Ga^{69}	20 min.	1.52	22.7	
As^{75}	26.8 hr.	4.2	23	
Br^{79}	18 min.	8.9	30 } 44	Isomeric ratio: Thermal 3.0; fast 2.1
	4.4 hr.	3.0	14 }	
Br^{81}	34 hr.	2.3	17	
Cb^{93}	6.6 min.	1.4	52	See text
Mo^{98}	67 hr.	0.39	32	
Mo^{100}	14.6 min.	0.22	15	
Rh^{103}	44 sec.	150	103 } 119.5	Isomeric ratio: Thermal 11.7; fast 6.3
	4.2 min.	12.8	16.5 }	
Ag^{107}	2.3 min.	48	127	
Ag^{109}	22 sec.	108	225	
In^{115}	13 sec.	57	63 } 243	Isomeric ratio: Thermal 0.36; fast 0.35
	54 min.	157	180 }	
Sb^{121}	2.8 day	6.8	90	
I^{127}	25 min.	6.8	107	
Ba^{138}	86 min.	0.56	2.6	
W^{186}	23 hr.	37	66	
Pt^{198}	31 min.	3.6	59	
Au^{197}	2.7 day	96	121	
Hg^{204}	5.5 min.	0.37	88	
Pb^{208}	3 hr.	0.00046	1.8	
Bi^{209}	5 day	0.015	3.0	

* Values taken from Seren's survey (see reference 4), or from unreported work at Argonne Laboratory.

II. EXPERIMENTAL METHOD

Unmoderated fission neutrons were obtained (Fig. 1) by irradiating a plate of uranium metal in a beam of slow neutrons from the thermal column of the Argonne heavy water pile. A foil placed inside heavy Cd near such a plate receives only unmoderated fission neutrons if all slowing down materials are carefully excluded. As the capture cross section varies as $1/E$, the presence of only a small number of moderated neutrons can vitiate the results. The flux of fast neutrons at the foil can be determined as accurately as the thermal flux can be measured, for the ratio of the fast fission flux to the thermal flux can be calculated from the geometry of the plate and the nuclear constants of uranium.

The procedure, in brief, is to activate a cadmium covered foil of the material of interest with fission neutrons while an identical bare foil is simultaneously activated with thermal (plus fission) neutrons. The two foils are then counted on a G-M counter.

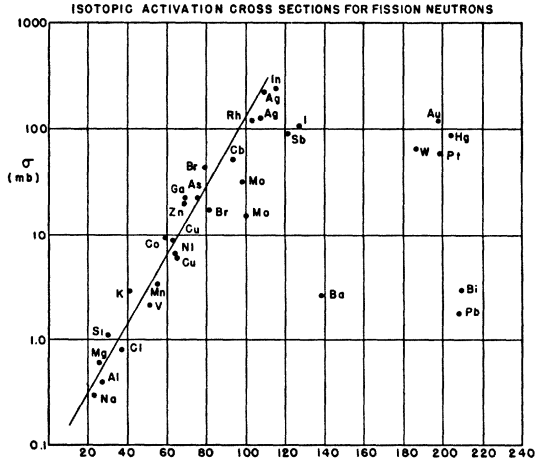

FIG. 2. Variation of measured isotopic activation cross sections (effective energy 1 Mev) with atomic weight.

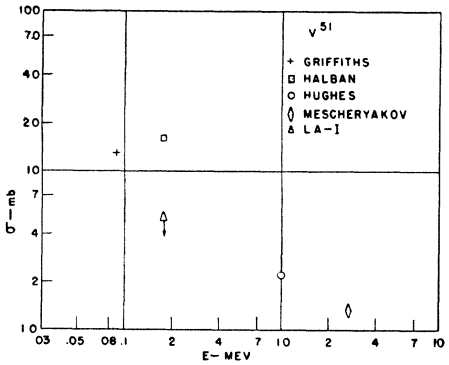

FIG. 3. Figures 3–17 are activation cross sections as a function of neutron energy for various isotopes. All available data are included, those of some observers being adjusted to obtain agreement for Au^{197}. See text for identification and description of the sources of data.

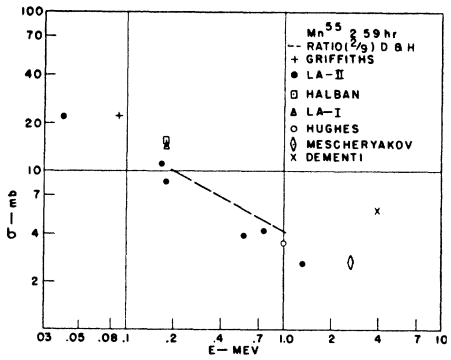

FIG. 4.

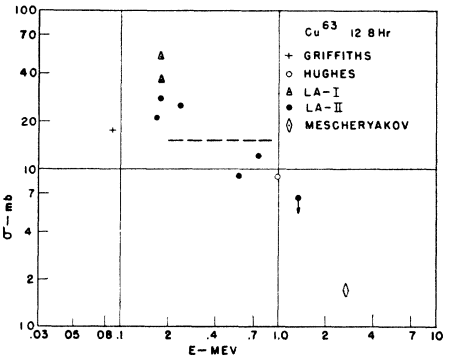

FIG. 5.

The fast activation cross section is obtained directly from the counting rate ratio, the known thermal cross section for the particular activity detected and the calculated ratio of fast to slow flux at the plate (which is approximately unity). In this way no corrections are necessary for self absorption in the foil, complicated decay schemes, or efficiency of the counter. In some cases where the thermal cross section was doubtful, it was determined from the bare foil activation, taking into consideration the thermal flux and the counter efficiency. Because the fast cross section is often only one-thousandth of the thermal it was found extremely important to use a thick, tight Cd cover for the fast irradiation to prevent significant leakage of thermal neutrons. A slight activation sometimes resulted from the small number of resonance neutrons present in the thermal column if the substance had a high resonance cross section. The resonance activation could be estimated easily, however, by running a Cd-covered foil with the uranium plates removed. The cross section obtained by this method is an average over the fission spectrum but, because the variation of $\sigma(n,\gamma)$ with energy is about the same for different elements near 1 Mev (approximately $1/E$), the measured cross section can be correctly considered as the capture cross section at 1 Mev neutron energy (as calculated from the known fission spectrum and a $1/E$ energy dependence).

The technique was first checked by measuring Au^{197}, I^{127} and In^{115} which have large activation cross sections which had already been measured as a function of energy. The results for these isotopes plotted at 1 Mev agreed well with the previous activation cross section values as is shown in Figs. 17, 15, and 14. From these first results it was concluded that the method gave the correct absolute capture cross sections at the calculated effective energy of 1 Mev. As the first elements had been done quite easily the measurements were then extended to elements of lower atomic weight for which no cross sections were available. The method proved quite powerful in measuring the light elements because the high flux made activations possible for extremely low cross sections. A rough survey soon showed: (1) a rapid decrease of the activation cross section, σ_{act} (isotopic), with decreasing atomic weight, A, for $A<100$; in this region σ varied nearly as A^4; (2) a rather smooth dependence of σ on A; the deviations from a smooth curve were not greater than the large experimental uncertainties associated with activation cross-section measurements.

It was seen that if the smooth dependence of σ_{act} (isotopic) on A for $A<100$ were true, then the total radiative capture cross section of an element for fast neutrons could be determined even if only one isotope of small abundance could be activated. This would be so because σ_a, total absorption, would be equal to σ_{act} (isotopic) for any isotope. The smooth dependence on A would also mean that the total absorption cross section of an element with no measurable activation could be predicted; thus it would be expected that iron would have a cross section of 6.5 millibarns (mb $=10^{-3}$ barn) at 1 Mev and would increase about as $1/E$ (see Section IV) for lower energies for at least several hundred kilovolts. The effect of structural materials and fission products in a "fast" pile could then be

predicted which is, of course, impossible for a thermal pile. Although the first measurements included only a few cross sections for heavy elements, it was obvious that the situation for $A>100$ was not simple, as Bi and Pb showed much lower cross sections than the other heavy elements, all of which were in the 100–200 mb range.

After the early measurements, which were undertaken mainly to aid in fast pile design, a great many more cross sections were determined because the method proved to be useful from the standpoint of basic nuclear theory. Recently, Alpher, Bethe, and Gamow[2] have shown an interesting relationship between the fast cross sections and the abundance of the elements and have interpreted the relationship in terms of a neutron-capture theory of the origin of the elements. The work was done at irregular intervals during the past several years but only the final results brought up to date will be presented here.

III. DISCUSSION OF RESULTS

Table I contains the final cross-section values for 32 isotopes, for 4 of which, both activities of isomeric pairs were measured. For each isotope the activity studied, its thermal isotopic activation cross section, and the fast activation cross section (which, barring unknown isomers, and to the extent that the isotopic cross sections follow a smooth curve will be equal to the atomic absorption cross section of the normal element, σ_a) are given. Figure 2 shows the cross sections plotted as a function of atomic weight where it is seen how the values for $A<100$ follow a rapidly rising smooth curve within a factor of about two. Thermal cross sections of course do not behave in such a regular manner and the curve of Fig. 2 can actually be used to infer peculiarities in the thermal cross section if the fast cross section (based on the thermal) does not fall on the curve. Examples of this behavior will be given among the following discussions of individual isotopes.

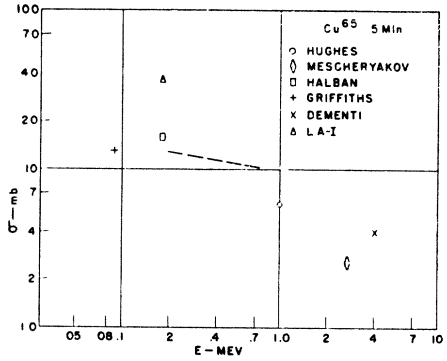

FIG. 6.

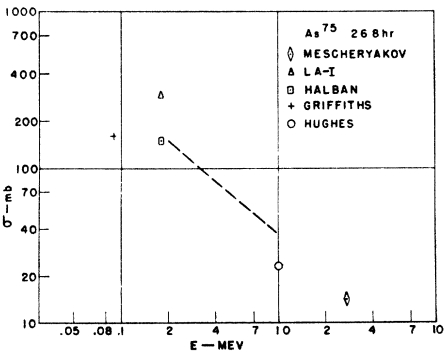

FIG. 7.

Cb93

At the time the present work was begun it was thought that the thermal activation cross section

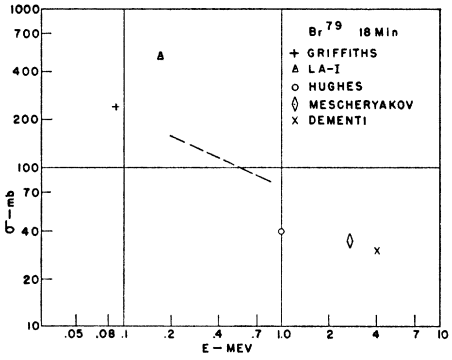

FIG. 8.

for 6.6 m Cb^{94} was only 0.01 b while the total thermal absorption (as measured by the effect on the pile) was known to be 1.4 b. The fast cross section measured in the present experiments, if based on a thermal value of 1.4 b, turned out to be 52 mb, which is "on" the smooth curve. This result implied that the total thermal activation cross section should be 1.4 b and not 0.01 b (i.e. that the difference was not caused by impurities as had been expected). The correctness of this procedure was shown when it was found by Goldhaber and Sturm[3] that Cb^{94} decayed by isomeric transition and the 0.01 b value was low by just the efficiency factor for γ-detection of a G-M counter, the correct activation cross section being about 1.4 barns.

Co59

With thermal neutrons, a strong 5 year and a 10.7 minute isomer are produced, with cross section in the ratio 41 (this ratio may actually be lower

[2] R. A. Alpher, H. A. Bethe, and G. Gamow, Phys. Rev. **73**, 1803 (1948); R. A. Alpher, Phys. Rev. **74**, 1577 (1948).

[3] M. Goldhaber and W. Sturm, Phys. Rev. **70**, 111 (1946).

because of uncertainty in the fraction of the 10.7 minute activity which grows into the 5 yr.). In the present experiments only the 10.7 minute activity was measured and the total fast activation cross section was obtained by multiplying the 10.7 minute isomeric cross section by 41. The agreement of the result, 9.2 mb, with the curve indicates that there is no large shift in isomeric ratio with neutron energy in this case.

Ni^{64}

The early results of the present method, based on the extant thermal cross sections, gave a fast cross section far off the curve. It was then decided to remeasure the thermal cross section. The resulting *atomic* activation cross section for the 2.6 hour activity was 0.030 b as compared to the old 0.017 b (Seren's[4] value). In the meantime it was shown by Swartout *et al.*,[5] that the 2.6 hour activity was Ni^{64} (0.88 percent) and not Ni^{62} (3.88 percent). The combined effect of these two corrections was to

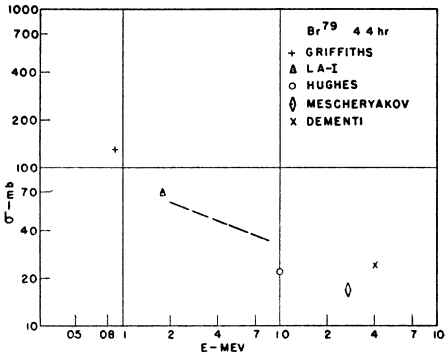

FIG. 9.

raise the fast isotopic cross section by a factor of about 7 and to bring it into agreement with the smooth curve.

Zn^{68}

Measurements of the isomeric activities (57 m and 13.8 h) again gave values off the curve when the listed values[4] of the thermal cross section were used. The thermal measurements were then repeated with the result that the reported 1.09 b isotopic value for the 57 m activity was lowered to 0.89 b and the 13.8 h lowered from 0.31 to 0.085 b. The new thermal cross sections led to a fast total activation of 20.1 mb again in agreement with the curve.

For the four isotopes for which both members of isomeric pairs were measured, the isomeric ratios

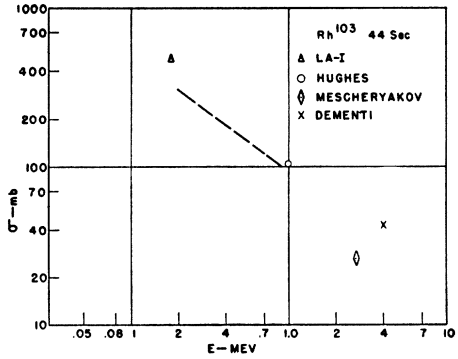

FIG. 10.

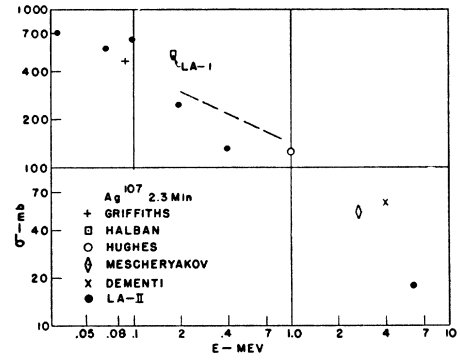

FIG. 11.

(ratios of activation cross sections for the isomers) for thermal and fast neutrons are also given in Table I. In the cases of In^{115}, Rh^{103} and Br^{79} the ratio is the same within experimental error for fast and for slow activation. For Zn^{68}, however, the ratio changes from a thermal value of 10.5 to 0.55 for fast neutrons. The fast isomeric ratio is of course the average of the ratio over many resonance levels of the compound nucleus and might be expected to be different (and closer to unity) than the thermal ratio which usually refers to a single level. The results, for the four isomeric pairs discussed although rough, are not inconsistent with this view as the changes which do show are in the correct direction. The behavior of Co^{59}, however, seems to indicate a large isomeric ratio which does not change greatly with neutron energy.

Among the heavy isotopes, there are several marked exceptions to the general rule that the cross sections are roughly constant, namely Ba^{138}, Pb^{208}, and Bi^{209}. Feshbach, Peaslee, and Weisskopf[1] had already pointed out that lead and bismuth, because of their low absorption, must have an unusually large level spacing D which would mean an unusually low neutron binding energy. However, as the same behavior in Ba^{138} was unexpected, attempts were made to find something wrong in the measurement or interpretation. Weisskopf sug-

[4] L. Seren, H. Friedlander, and S. Turkel, Phys. Rev. **72**, 888 (1947).

[5] J. A. Swartout, G. E. Boyd, A. E. Cameron, C. P. Keim, and C. E. Larson, Phys. Rev. **70**, 232 (1946).

gested that perhaps competition by inelastic scattering caused the low cross section in Ba^{138}. This possibility was checked by measuring the neutron capture cross section with Na-Be (900 kev) and Na-D_2O (230 kev) photo-neutrons. The cross section was unusually low, however, even at 230 kev where no inelastic scattering would be present. In the meantime Mayer[6] had shown that certain numbers of neutrons in nuclei formed stable "shells" and that the binding energy for the next neutron is extremely low. The low cross section for Ba^{138} was then easily explained because it contains 82 neutrons, forming a closed shell (Pb^{208} and Bi^{209} each contain 126 neutrons, also a closed shell).

IV. COMPARISON WITH OTHER MEASUREMENTS

As already discussed, the results for Au, I, and In were compared with the existing data on cross sections *vs.* energy as a check on the general method before proceeding with the light elements. After the other measurements had been made, an exhaustive survey by Way and the isotope information group at Clinton showed that there were quite a number of old results on capture cross section for neutrons of various energies with which the present results could be compared. In many cases the energies were not well defined and the absolute cross-section values were not good. However, if the cross sections of a given experimenter are multiplied by a constant to bring his value for a standard substance, say Au^{197}, into agreement with Fig. 17, then a comparison of the different values is possible. It is assumed in this procedure that the relative values of a given observer are correct even when his reported absolute values are definitely wrong.

Curves based on all the available data for a number of isotopes are given in Figs. 3–17. All those isotopes are shown for which more than 3 points were available above 0.1 Mev. The sources of data, as marked on the figures, are as follows:

"DEMENTI": V. S. Dementi and D. V. Timoshuk, Comptes Rendus de l'Acadamie des Sciences de l'URSS **27**, 929 (1940). Activation measurements using a Rn-Be neutron source, with a reported predominant energy of 200 kev. The reported cross sections are much too low if plotted at 200 kev and it seems more reasonable to assume an energy of about 4 Mev (as K. Way has done in her compilation) for which value the results are in line with the other measurements.

"D & H" (dashed line): P. Demers, H. H. Halban, R. Allen and G. R. Bishop. Nature **161**, 727 (1948). Measurements of the ratio of the cross sections at 220 kev (ThC″-D_2O photo-neutrons) to those at 950 kev (ThC″-Be). Only the ratio was measured, which is shown by the slope of the dotted line, the ordinate being arbitrary.

"WATTENBERG": R. Fields, B. Russell and A. Wattenberg: Unpublished activation measurements using Na-D_2O (230 kev) and Na-Be (900 kev) photo-neutron sources.

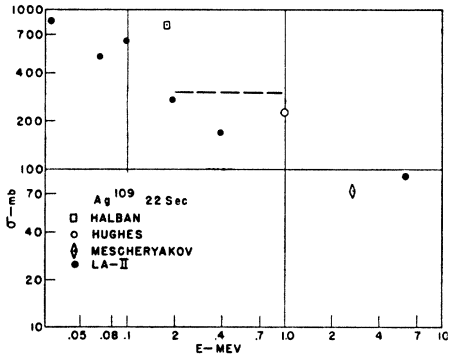

FIG. 12.

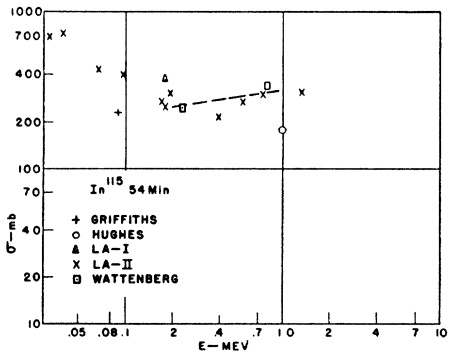

FIG. 13.

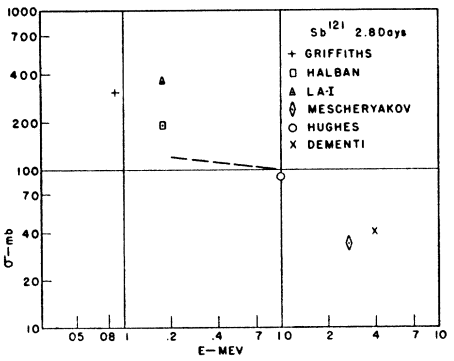

FIG. 14.

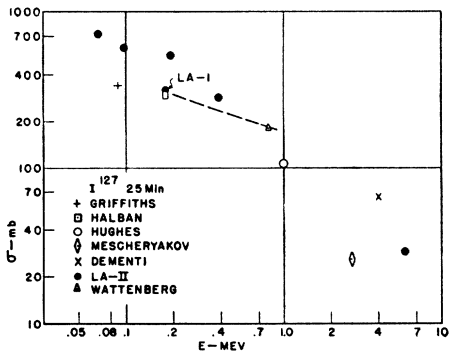

FIG. 15.

[6] M. G. Mayer, Phys. Rev. **74**, 235 (1948).

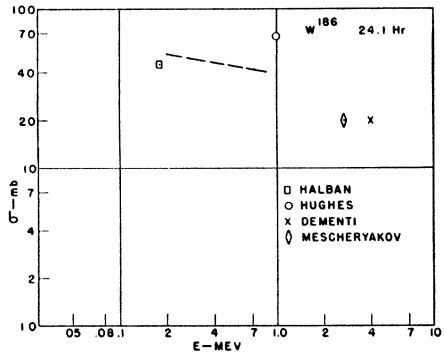

FIG. 16.

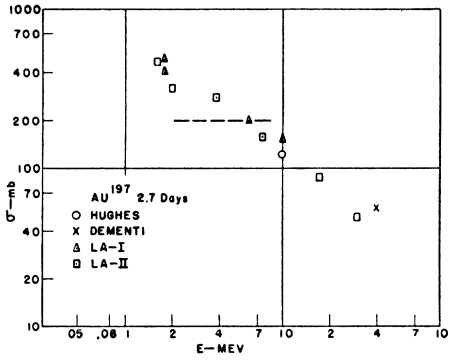

FIG. 17.

The cross sections were measured relative to gold, taking the value for gold from the data of Fig. 17.

"GRIFFITHS": J. H. E. Griffiths. Proc. Roy. Soc. **A170**, 513 (1939). Cross sections measured with a Ra-γ-Be source, using a calculated neutron flux from the source for obtaining absolute cross-section values. In plotting Griffiths points, we have followed K. Way in selecting 90 kev as the effective neutron energy which means that the cross section must be multiplied by 4.9 to fit the Au curve.

"HALBAN": H. H. Halban and L. Kowarski: Nature **142**, 392 (1938). Used ThC″-D_2O photo-neutron source of energy 220 kev. It was necessary to multiply the cross sections by the surprisingly large factor of 17 to secure agreement with the curve for gold given in Fig. 17.

"HUGHES": Present results.

"MESCHERYAKOV": M. G. Mescheryakov. Comptes Rendus URSS **48**, 555 (1945). Activations measured using $D(d,n)$ cyclotron neutrons of energy 2.7 Mev. Reported cross sections have been multiplied by 0.13 to agree with Fig. 17.

"LA-I": Photo-neutron measurements made at Los Alamos by:

M. Deutsch, M. B. Fischer, S. L. Friedman, A. O. Hanson, C. D. Klema, G. A. Linenberger, M. L. Perlman, E. Segrè, J. H. Williams

"LA-II": Van de Graaff measurements made at Los Alamos by:

J. M. Blair, M. Deutsch, K. I. Greisen, A. O. Hanson, G. A. Linenberger, J. A. Miskel, R. F. Taschek, C. M. Turner, J. H. Williams

The curves of Figs. 3–17 show that once the absolute cross-section values are adjusted for a given isotope there is quite good agreement for the others. The variation of cross section with energy is quite similar for most of the isotopes, that is, approximately a $1/v$ dependence at low energies which changes over to $1/E$ in the region of several hundred kilovolts. This change in slope would be expected to take place where the neutron width becomes greater than the gamma-width and its location in the experimental curves is quite reasonable. The continuation of the $1/E$ slope to energies well above one Mev indicates that the increase in capture (relative to $1/E$) caused by high l values must be approximately compensated by the decrease caused by inelastic scattering. The only definite exception to the general behavior seems to be In^{115} (and to a smaller extent, Ag^{109} and Sb^{121}) which changes much more slowly with energy than the other isotopes.

The measurements are being continued at the present time with special reference to the "magic number" nuclei containing 50, 82 and 126 neutrons. The unusually low binding energy for the next neutron of these nuclei should be shown quite well in their fast neutron capture cross section because of the rapid variation of level spacing with excitation energy.