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January 12, 1951

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January 12, 1951

Abstract

Relative photo-neutron yields were determined for 50 elements bombarded by the 330 Mev x-ray bremsstrahlung of the Berkeley synchrotron. The determinations were made at 90° to the beam axis with a BF₃ proportional counter surrounded by a paraffin moderator. Total relative errors were between 4 percent and 12 percent. Absolute yields per unit beam energy were determined using an Argonne calibrated Ra-Be neutron source, and the beam energy measurements of Blocker and Kenney. Relative yields for the elements above Z = 30are proportional to $Z^{1.7}$. Yields for elements of low Z show a correlation with the binding energy of the last neutron. Angular distributions of photoneutrons were briefly investigated, and a Pb transition curve for photo-neutrons was run. RELATIVE PHOTO-NEUTRON YIELDS FROM THE 330 MEV BREMSSTRAHLUNG

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Radiation Laboratory, Department of Physics University of California, Berkeley, California

January 12, 1951

Previous to July, 1950, workers at the General Electric¹ laboratories had. determined yields from some (γ, n) , $(\gamma, 2n)$, and (γ, np) reactions by studying the induced activities. Kerst² had studied relative neutron yields for over 50 elements from x-rays of the 22 MeV Illinois betatron by neutron counting. Strauch³ had studied a few reactions by induced activities, and with transition curves had determined the reaction energies for these reactions, finding resonant energies as high as 80 MeV.

In view of Strauch's findings, a comprehensive study was made of photoneutron yields from the 330 MeV x-ray bremsstrahlung by a direct neutron counting experiment.

Briefly, the experiment here described consisted of bombarding various elements with the x-ray beam of the synchrotron and counting the ejected photoneutrons in a direction 90° to the beam axis with a BF₃ neutron counter.

Experimental Procedure

The boron trifluoride proportional counter filled to one atmosphere with 90 percent B^{10} boron trifluoride, was 10 inches in length, 1 1/4 inches inside diameter, and in a 1/8 inch brass envelope, with a 3/16 inch brass cap over the front end. This counter fed its output directly into a cathode follower mounted in the same brass case that held the counter.

The cathode follower output was fed into the standard UCRL 125 ohm linear amplifier input. Its output was fed to the input of a scaler. The counter tube was operated at 2300 volts. The combination of counter voltage, amplification, and discriminator setting was chosen from runs with the 5 mg Ra-Be neutron source and a pure Ra source (of gamma rays only) as optimum for high neutron counting efficiency and complete discrimination against gamma rays and noise. The counter was found to vary slowly in sensitivity over a period of hours.

In order to count high energy neutrons, this counter was imbedded in a cylinder of paraffin 8 inches in diameter and 10 inches long with a 1 1/2 inch hole along the axis for the counter. This cylinder was contained in a 1/8 inch brass cylinder with a 62 mil cadmium sheet over it. Outside this was another 3 inches of paraffin and the entire structure was contained in a brass case which could be moved easily by an overhead crane. A cross section of the counter showing its geometry is given in Fig. 1. This geometry is given in Rossi and Staub⁴ as having a uniform response to neutrons of all energies to 5 Mev, with the sensitivity dropping perhaps 20 percent at 5 Mev. Kerst* mentions that his laboratory has estimated that a similar counter is 60 percent as efficient for 15 Mev neutrons as for low energies.

A check showed that the counting rate increased slightly as the Ra-Be neutron source was moved perpendicularly off the axis of the counter. This would introduce an error of 3 percent into the data for targets 3 inches broad.

At 7 inches from the end of the counter, it was found that the neutron counting rate varied as the reciprocal of the distance, so that no corrections were made for the finite extent of targets and beam along the counter axis.

The x-ray beam was collimated to $1 \frac{3}{4}$ inches diameter at the position of the targets studied. These targets were all of such a size as to entirely

* D. W. Kerst, private communication

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contain the x-ray beam. Collimation was accomplished by a properly tapered hole in a 9 inch lead wall placed 5 feet from the 20 mil platinum x-ray target in the synchrotron. The beam was monitored by an ionization chamber placed on the synchrotron side of the collimator. This was connected in such a way as to integrate the total beam energy. It was calibrated by Blocker and Kenney⁵ for the various diameter collimators used, so that it was possible to determine absolute neutron yields per erg of beam energy.

The targets were place 20 inches outside the lead collimator. Since all were large enough so that the x-ray beam was entirely within them, only the thickness and density of the targets was important. The targets used consisted of metal plates, discs, or slabs of the element where such was available. For nonmetallic elements, crystals or powders of the elements were contained in one of three lucite containers. Other elements, such as Cl, F, La, etc., were available only as compounds, so powders or crystals of these compounds were used, placing them in the lucite containers.

The x-ray beam is partly absorbed and partly transformed in passing through a finite thickness of a target sample according to known shower theory. From a transition curve taken from a lead target it was found that the neutron yield was reduced 3 percent if the target was one-quarter shower unit thick instead of infinitesimally thin, or 4 percent for a target one-third shower unit thick. All targets were kept less than one-third shower unit thick, and most were less than one-quarter unit thick. The computation used was based on the fact that 1 s.u. in lead is 0.5 cm, and the thickness equal to 1 s.u. for an element is proportional to $\frac{1}{Z^2}$. $\frac{1}{Z^2N} \approx \frac{1}{ZN}$

To determine the number of moles per cm² of each target, it was weighed and measured to three significant figures. Metallic targets were all rectangular or square, therefore for them, $\frac{\text{moles}}{\text{cm}^2} = \frac{\text{wt.}(\text{gns})}{\text{area}(\text{cm}^2) \times \text{M.W.}}$. The substances in powder or crystalline form were poured into the appropriate lucite container until it was filled to a certain line. The area of the container below this line was carefully measured. Then the weight of the container was subtracted from the weight of powder plus container to determine moles/ cm^2 . The error in moles/ cm^2 for metallic targets was probably less than 2 percent, for powders and crystals the error was possibly as high as 5 percent due to irregularities of the substances. Moles/ cm^2 for some targets, such as water and CCl₄, were computed from handbook densities and the container thickness directly.

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The metallic targets and lucite holders were held in the synchrotron beam by a brass structure with a platform of adjustable height on which the targets were placed. The bulk of the structure was over a foot below both the beam axis and the counter axis. This minimized gamma and neutron scattering problems. Provision was made on this holder for centering the targets accurately in the beam to within 1/8 inch. X-ray films were exposed in the beam at the time of each experiment run to check the beam centering.

Neutron shielding of the counter from the stray neutron flux in the magnet room was accomplished with 12 one hundred pound boxes of paraffin, plus two hundred pounds of smaller pieces of paraffin. In addition, a one foot cube of paraffin with a 4 inch hole in it was placed immediately after the lead collimator. With this paraffin, the counter was shielded completely with at least 12 inches of paraffin all 'around except for a passageway along the x-ray beam axis to allow access to the target.

A cadmium "hood" was placed over the front of the counter and another piece of cadmium placed behind the target so that the front of the counter "saw" only cadmium backed by paraffin. This geometry decreased the background by a factor of three over what it had been using only paraffin shielding directly between the counter and the lead collimator. An overhead view of the experimental -7-

arrangement is shown in Fig., 2.

Actual runs for data were made by running the beam from one to four or five "nunans" (a unit on the beam integrator) on each target so that a total of 2000 - 3000 neutron counts was obtained for each target. From this, statistical counting errors were \sim 2 percent to 10 percent. At the beginning, and after each group of from three to six targets, the tungsten target was run as a check on the stability of the counter. Then the ratio of the neutron counts per nunan from the tungsten target to the neutron counts per minute from a 5 mg Ra-Be source placed in the target position was accurately found. This 5 mg uncalibrated Ra-Be source was compared with a standard 500 mg Ra-Be source using the BF3 coun- $\frac{\text{neutrons}}{\binom{\text{mole}}{\text{cm}^2} \mathbf{x} \text{ erg}}$. The 500 mg ter, enabling reduction of data from $\frac{\text{counts}}{(\frac{\text{mole}}{\text{cm}^2})x}$ erg to source had been calibrated to within 5 percent by comparison with an Argonne Laboratories standard. From these calibrations it was found that the BF_{3} counter in the above described paraffin geometry had an efficiency of 0.35 percent for Ra-Be neutrons entering the 8 inch diameter face from a point on its axis 7 inches from the end of the counter. The data were taken at this distance.

A check was made to see whether neutron scattering in the target had any appreciable effect on the yield. An angular distribution run was made on a normally isotropic photoneutron emitter, tungsten. This target was a flat plate 0.120 inch thick and 2.5 inches wide. Yields were taken with the counter axis at 90° to the beam axis and the plane of the target at 90° and 120° , then with the counter at 120° and with the target plane again at 90° and 120° . The counter was 20 inches from the target. These data are given heres:

counter axis angle	target plane angle	neutron yield (rel.)
90 ⁰	900	9,4
, 90 ⁰	120 ^{0.}	16.4
120 ⁰	900	16.4
120°	120 ⁰	9.7

It is evident that there is appreciable neutron scattering if the neutrons have to traverse the entire target width before reaching the counter. Consequently, the targets were tipped with their plane faces at 60° to the beam axis so that the neutrons reaching the counter would have to go only through the target thickness and not through its breadth.

The synchrotron gives a pulse of x-rays 3000 microseconds long with six pulses a second. By studying the counts per nunan as a function of beam intensity and observing the pulses on the synchroscope, the resolving time of the counter was determined to be about 3 microseconds. This gave rise to less than 1 percent error if the neutron counting rate was held to less than 3200 counts per minute.

At all times during experimental runs, the output of the linear amplifier was monitored with a synchroscope. This gave reassurance of the synchrotron x-ray pulse length, consistency of counter characteristics (by observing the a-particle pulse shapes), and resolving time.

The scalar was gated to the x-ray beam pulse, although this was found to be unnecessary except for operation with very low beam intensity. The background between pulses amounted to less than 10 counts per minute,

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The data are presented in $\frac{\text{neutrons}}{(\frac{\text{mole}}{\text{cm}^2}) \times \text{erg}}$. This is equivalent to saying that the data represent the number of neutrons that would be given off if one erg of bremsstrahlung passed through one mole of the target spread uniformly over an area of one square cm normal to the x-ray beam. This yield is also equal to $N_0 \int_0^{340} \sigma(w) f(w) dw$, where $\sigma(w)$ is the photo-neutron cros's section, w the photon energy, N_0 Avogadro's number, and $f(w) = \frac{dq}{dw}$, the number of quanta per unit energy range for one erg of beam. $(\int_0^{340} w f(w) dw = 1 \text{ erg.})$

The final data are given in tabular form in Fig. 3, on a log-log graph in Fig. 4, and on a semilog graph in Fig. 5.

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Relative yields may be in error due to various sources as follows:

comparison with W target	2%
$moles/cm^2$ determination	2 to 5%
reading beam integrator	2%
finite lateral extent of target	1%
beam degradation in target	2%

statistical fluctuation in counts 2 to 10% This gives a probable error of 4 percent to 10 percent in relative data.

From Fig. 4 the relative photo-neutron yields of the elements show that the yield varies as $Z^{1.7}$ for Z greater than 30. Present theory^{*} favors a $Z^{1.5}$ dependence or even a lesser slope. Kerst, and Baldwin and Elder⁶ have reported a Z^2 variation.

For the low Z elements, an even-odd alternation is seen. This is roughly related to the binding energy of the last neutron, as may be seen from the plot of yield/A vs. the binding energy of the last neutron in Fig. 6. This graph appears as a straight line for 2 < Z < 22.

Deuterium and beryllium show abnormally high yields on the yield vs. Z plot probably due to the very low binding energy of their last neutron. Uranium and thorium yields are high due to photofission neutrons adding to the usual photo-neutrons.

Kerst's results at 22 Mev are also shown in Fig. 5 normalized to our Pb value. These points show that for low Z elements, the photo-neutron cross sections have appreciable values above 22 Mev. The 22 Mev data are seen to duplicate the 330 Mev even-odd alternation.

The absolute data in $\frac{\text{neutron}}{(\frac{\text{moles}}{\text{cm}^2}) \text{ x erg}}$ is 1.27 times the yield found by Kerst

* H. Bethe, private communication

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using the 320 Mev Illinois betatron. This latter data appeared after the Berkeley investigation was partially complete, as the result of a brief investigation by Kerst.⁷

The estimated error in yield in absolute units is due to the following: absolute beam calibration (Blocker and Kenney) . . . 10% calibration of 500 mg Ra-Be standard neutron source . 5% calibration of 5 mg source from 500 mg source . . . 2%

The table of the data includes a column giving the counting error and the total relative error for each element. The relative error includes all errors other than absolute beam and counter calibration. The validity of using compounds is demonstrated by the data for the alkali halides, which provide cross checks on Na, F, and K. An earlier experimental run, before the target tipping effect was found, had given the yield of nitrogen from liquid nitrogen 11 percent less than the yield derived from an NH_4NO_3 target. Due to constant boiling of the liquid nitrogen, it was therefore considered that NH_4NO_3 was a fair source of nitrogen data. It was found that elemental carbon gave a neutron yield only 1.0 percent less per mole than polyethylene (CH_2), so that it was thereafter considered safe to assume the photo-neutron yield of hydrogen to be zero. Therefore the oxygen data were taken directly from water, and the deuterium data were found by subtraction of the yield from a container filled with water from the yield of the same container filled with D₂O.

Angular Distribution

A brief investigation into the angular distribution of the photo-neutrons was made. This was done by suspending the counter from an overhead crane and looking at the targets with it at various angles from the beam axis. For these As Constants

investigations the targets used were cylinders of the elements investigated to minimize the effects of non-uniform scattering of neutrons by the target. For angular distribution measurements, beam degradation was a second order effect, even though some targets were over a shower unit in diameter to increase counting rate. The targets were placed 150 inches from the lead collimator, and the counter was placed with its end 15 inches from the target center. Paraffin shielding was placed only between the counter and the synchrotron, and between the counter and the floor, Data were taken at 389, 90°, and 142° to the beam

The results, plotted in Fig. 7, show that photo-neutrons are given off with spherical symmetry except for deuterium, carbon, and sulfur. Therefore, except for the lightest elements, the 90° photo-neutron data are valid for total yields. The probable counting errors were 20 percent to 25 percent for the deuterium target, 10 percent for the carbon and sulfur targets, and less than 5 percent for the heavier elements. Some slight increase in yields was seen at small angles (30°), for some targets, but this appeared due to x-rays scattered by the target sample producing neutrons in the counter itself.

Transition Curves in Lead.

With the geometry used for the relative photo-neutron yields, a set of data was taken for determination of a lead transition curve. This was done by using lead squares of varying thickness as targets. The lead squares were all 2 1/2 in. x 2.1/2 in. square, and varied from 0.020 inch to 2.3 inches: (11) shower units) in thickness. The thicker blocks were made by stacking thinner squares together. Neutron counts were taken for each thickness, then the data were reduced by dividing counts by thickness to give an integral transition curve. This was plotted as <u>counts</u> vs. thickness and a smooth line drawn

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connecting points. Then this smooth curve was "differentiated" by determining the difference in counts per thickness in unit thickness. The new points were then plotted against thickness which give the true transition curve shown in Fig. 8. This corresponds to the neutron yield a unit foil of lead would give when placed behind the abscissa thickness in lead.

There is an error introduced into the curve at large thicknesses due to the finite diameter (2 1/2 inches) of the lead target. Blocker and Kenney found a large angular spread in the showers they studied.

This technique is not applicable to light elements, since it was found experimentally impractical to reduce the background from the showering lead to the point where the neutrons from even a large sample of the light element could be distinguished.

However, the transition curve obtained for lead shows interesting properties. The initial sharp descent found by Strauch to correspond to an 80 Mev resonance is seen; but this is followed by a broad rise peaked at about 4 s.u. which apparently indicates a resonance at 15-20 Mev.

We wish to express our gratitude to Professor A. C. Helmholz for his guidance throughout this experiment.

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This work was performed under the auspices of the Atomic Energy Commission.

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Fig. 1



Fig. 2

Fig. 3

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Element	Z	Form	Counting Error %	Total Error (Relative) %	neutrons mole/cm ² erg
D	 		A .	(1102/002/00) //	
ر • •	7		4	7	10.0
Li	. 3	M	3	5	10.7
Be	4	M .	2	4	15.2
В	5	B ₄ C pdr	3	7	13.3
C	6	M	3	5	9.08
N	7	NH ₄ NO ₃ pdr	7	9	15.7
0	8	H ₂ O liq	3	7	13.3
F	9	LiF pdr	6	9	28.7
		NaF pdr	6	9	26.8 27.9
·		CaF2 pdr	7	9	28.1
Na	11	NaCl pdr	7	9	27,8
		M	3	5	29.7
Mg	12	M	2	5	22.8
Al.	13	M	2	5	32.2
Si	14	pdr	5	8	22.4
Р	15	pd r	4	7	45.3
S .,	16	pdr	3	7	23.4
Cl	17	CCl ₄ liq	2	6	49.6
K	19	KCl pdr	6	9	43.6
	, ,	KF pdr	3	7	52.0
Ca	20	M turnings	3	7	31.4
Ti	22	TiO2 pdr	4	7	99 .2
Δ	23	V ₂ O ₂ Cl ₄ pdr	8	10	90,5

Fig. 3 (Cont.)

Table of Results

			Counting	Total Error	neutrons	
Element	Z	Form	Error %	(Relative) %	mole/cm ² erg	
Cr	24	pdr	2	6	107.7	
Mn	25	flakes	2	6	143.2	
Fe	26	M	· ′· 2	4	115.5	
Co	27	Co ₂ 0 ₃ pdr	10	12	170	
Ni	28	M	2	4	85.4	
Cu	29	M	2	4	173	
Zn	30 ₍	М	2	4	220	
As	33	pdr	2	6	236	
Se	34	pd r	2	6	283	
Br	35	NaBr pdr	2	6	267	
Sr	38	SrCl ₂ pdr	4	7	368	
Zr	40	Zr02 pdr	2	6	324	
No	42	M	2	4	368	
Ag	47	Μ	2	4	422	
Cđ	48	M	2	4	428	
In	49	M	3	7	555	
Sn	50	M	2	4	503	
Sb	51	pdr	2	6	610	
I	53	crystals	2	6	578	
Ba	56	BaO pdr	_ 2	6	606	
La	5 7	$La_2(C_2O_4)$.	4	7	805	
Ce	58	9H20 par CeO ₂ par	2	6	695	
Ta.	73	Μ	2	4	998	
W	74	M	2	4	1051	
Pt	78	M	2	4	1110	

Fig. 3 (Cont.)

Table of Results

Element	Z	Form	Counting Error %	Total Error (Relative) %	<u>neutrons</u> mole/cm ² erg
Au	79	M	2	4	1134
Hg	80	HgC1 pdr	2	6	1073
Pb	82	M	2	4	1140
Bi	83	M	2	4	1185
Th	90	Μ	2	4	1980
U	92	M	1	4	2170



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Fig. 4



Fig. 5



Fig. 6







Fig. 8