Yields and Angular Distributions of Some Gamma-Neutron Processes*

G. A. PRICE AND D. W. KERST
University of Illinois, Urbana, Illinois
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Measurements of $\gamma-n$ yields for 53 elements have been made using betatron bremsstrahlung with maximum energies of 18 Mev and 22 Mev. The neutron flux was observed by a rhodium detector and compared with a standard Ra-Be neutron source. In the region from $Z=22$ to $Z=83$ the $\gamma-n$ yield in neutrons per mole per r of radiation is observed to be 2270. $Z^{2.1}$ when the bremsstrahlung spectrum has a 22-Mev maximum energy. The regularity of neutron yield as a function of atomic number indicates a definite similarity in the $\gamma-n$ cross-section curves for neighboring heavy elements. The trend of the observed yields is in rough agreement with the yields predicted by Goldhaber and Teller. Estimates are made of the fractions of thorium and uranium atoms which undergo photo-fission.

Angular distributions of photo-neutrons using bremsstrahlung with a 19.7-Mev maximum energy show no spherical asymmetry greater than the probable error for iron and lead whereas deuterium and beryllium have maxima at 90°.

MOST measurements of $\gamma-n$ cross sections have required the detection of a beta-activity following the emission of a neutron. Previous experiments can be classified according to the type of gamma-rays used. Wäffler and Hirzel[1] have measured the relative $\gamma-n$ cross section for 35 isotopes using the 17.5-Mev gamma-ray from the lithium (p,γ) reaction. High energy bremsstrahlung from betatrons have been used by McElhinney[2] and by Friedlander and Perlman.[3] These cross-section measurements have been restricted to those isotopes which have a detectable beta-decay following the $\gamma-n$ reaction.

An alternate method for cross-section measurements is to measure the number of neutrons emitted in the $\gamma-n$ process. This has been done at low gamma-energies in measuring the angular distribution of neutrons[4] from light elements. A similar method is described below for measuring the absolute neutron yields and angular distributions.

Bremsstrahlung from the betatron with maximum energies of 18 Mev and 22 Mev were used for the absolute $\gamma-n$ yields and 19.7 Mev for angular distributions. Samples consisted of natural isotopic mixtures, usually of a single element. The only enriched sample was deuterium oxide. Some of the samples were chemical compounds in which cases corrections were made for the presence of undesired elements. Most of the samples were chemical reagents with a high degree of purity. Sample weights varied from 54.5 grams to 567 grams.

The detector was a rhodium foil in a large paraffin block placed in a double wall Lucite box. The $\frac{1}{4}$-in. space between the walls was filled with B_4C. Fast neutrons from the irradiated sample pass through the B_4C, are moderated by the paraffin, and as slow neutrons are captured by the rhodium (Fig. 1). The 44-second beta-activity in rhodium is observed with a Geiger counter. The standard procedure was to irradiate the sample for three minutes, wait 30 seconds, and measure the Rh beta-activity for three minutes. Slow neutrons from the room were absorbed by the boron shield. Large paraffin blocks shielded the detector on all sides except the side facing the sample. A 24-inch concrete wall in front of the betatron shielded the detector from x-rays and also helped moderate the fast neutrons from the betatron.

A rhodium foil placed in a cavity of the concrete wall served as the x-ray monitor. The wall moderates fast neutrons from the betatron. Since the number of neutrons generated in the betatron is proportional to the x-ray intensity and is much greater than the number produced in the sample, the beta-activity of the monitor Rh foil is a good measure of the x-ray intensity. Since the detector and monitor integrate the neutron intensities with the same 44-second decay period, fluctuations in x-ray intensity do not have to be considered.

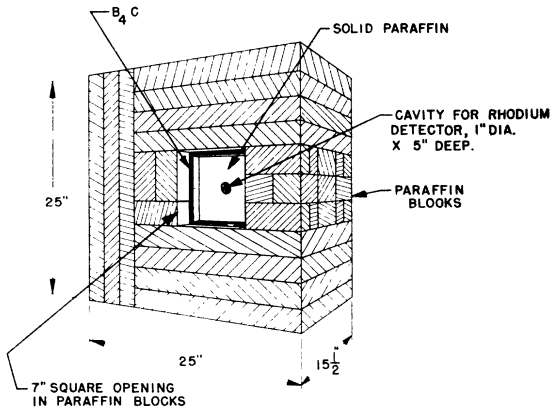

FIG. 1. Fast neutron detector. The front plate, which is made of Lucite and boron carbide, has been removed to show the location of the rhodium detector.

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[1] Wäffler and Hirzel, Helv. Phys. Acta, **21**, 200 (1948).
[2] J. McElhinney *et al.* (not published).
[3] G. Friedlander and M. L. Perlman, Phys. Rev. **74**, 442 (1948); Phys. Rev. **75**, 988 (1949).
[4] Hammermesh, Hammermesh, and Wattenberg, Phys. Rev. **76**, 611 (1949).

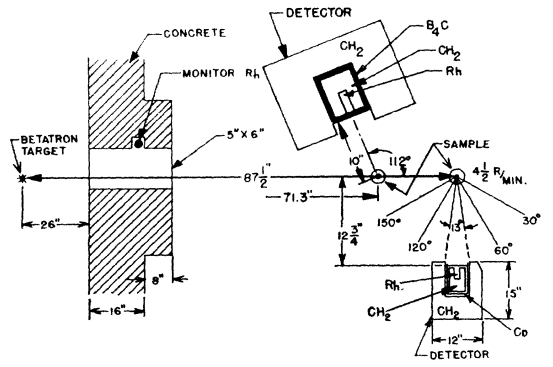

FIG. 2. Schematic diagram of apparatus. During measurements only one detector was in position at any time.

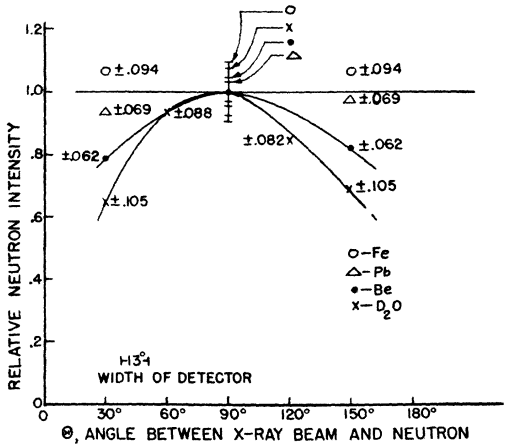

FIG. 3. Angular distributions of photo-neutrons for bremsstrahlung spectrum with maximum energy of 19.7 Mev. Intensities are normalized at 90° and probable errors are given for each point.

A 25-mg standard radium-beryllium neutron source was used to calibrate the neutron detector by placing it at the normal sample position. The x-ray monitor was calibrated by putting a thick walled Victoreen ionization chamber at the normal sample position.

Neutron background was measured by removing the sample completely from the x-ray beam. The sample was supported by fine wires to minimize the background.

Corrections for x-ray absorption in the sample were made assuming monochromatic x-rays for 13.73 Mev, using absorption coefficients by Adams.[5] Interpolations were made for those elements whose absorption coefficients had not been measured. The absorption correction factors varied from 1.00 to 1.97. This is the factor by which the observed neutron yield was multiplied to get the true neutron yield. If the absorption coefficient for 19.1 Mev is used instead, the corrected yields are changed by $5\frac{1}{2}$ percent for Hg and Mo which are the extreme cases but generally the change is less than 2 percent.

Probable errors were calculated on the basis of the counts registered by the Geiger counters. Each sample was tested at least twice and the variation in the neutron yields was generally within the statistical fluctuations to be expected on the basis of total counts.

There had been some indication that the neutron distribution from $\gamma-n$ processes was not spherically symmetrical. A rough test of this distribution was made for lead, iron, beryllium, and deuterium. The deuterium test with a 100-gram sample of heavy water was merely for the purpose of proving that asymmetries could be observed.

Neutron intensities were measured at various angles relative to the x-ray beam by a smaller detector which could be moved around. The detector subtended an angle of 13° from the sample, and could be moved from 30° to 150° with respect to the x-ray beam (Fig. 2). The probable error in the angular distribution values is relatively high because the small detector gave less activity in the rhodium foil and a lower sample to background ratio. The small detector had cadmium shielding, whereas the large detector had boron carbide shielding.

Neutron intensity distributions from deuterium and beryllium have a definite asymmetry with a maximum at 90°. In the cases of iron and lead any asymmetry is less than the probable error of the measurements. Figure 3 shows the angular distributions of neutrons from the four samples where they have been normalized at 90°. The probable error for each sample is also shown.

Figure 4 shows the absolute neutron yield expressed in neutrons per mole per r of radiation plotted against atomic number. The two sets of points represent measurements with the betatron operating at a maximum energy of 18 Mev and 22 Mev. The solid curves in the range from Z equals 22 to 92 show the best fit of the function aZ^b to the data. Between 82 and 92 the solid curves are supplemented by dotted curves which pass through the thorium and uranium yields. Below $Z=20$ curves have been drawn to indicate general trends only. In the region of low atomic number it appears that elements of odd and even atomic number behave quite differently. Probable errors are indicated by a vertical line through each point.

Figure 5 shows the gamma-neutron yields which have been obtained by McElhinney, Wäffler, and Hirzel, and Friedlander and Perlman. Neutron yields by McElhinney are absolute, whereas the other yields are relative and have been normalized at silver. These yields are due only to one isotope in each element, or in the cases where two isotopes of an element were used a weighted average was taken.

DISCUSSION

Two of the deviations of neutron yield from the general trend are for thorium and uranium. In each case the neutron yield seems to be much higher than the yield one would expect by extrapolating the curve found at lower atomic numbers. From these abnormally

[5] G. D. Adams, Phys. Rev. 74, 1707 (1948).

high yields one can estimate the fraction of uranium atoms or thorium atoms which undergo photo-fission instead of emitting photo-neutrons.

The ratio, R, of the actual neutron yield for uranium to the extrapolated photo-neutron yield is 1.76 at 22 million volts and 1.64 at 18 million volts; while this ratio for thorium at 22 million volts is 1.36, and at 18 million volts it is 1.26. If the fraction of excited nuclei which undergo photo-fissions is K, and if we assume that the number of neutrons produced per fission is between two and three, say 2.5, then taking the extrapolated yield as the measure of the number of nuclei excited,

$$2.5\times K+(1-K)=R.$$

Solving for K, we obtain

$$K=(R-1)/1.5.$$

This branching ratio, K, is 0.51 for uranium bombarded by 22-million volt x-rays, 0.43 for uranium bombarded by 18-Mev x-rays, 0.24 for thorium bombarded by 22-million volt x-rays, and 0.17 for thorium bombarded by 18-million volt x-rays. Thus, about one-half of the disintegrating uranium atoms disintegrate by photo-fission and about one-fourth of the thorium atoms disintegrate by photo-fission.

The observed photo-neutron yields show such regularity as a function of Z that it is possible to make a comparison with theory. Goldhaber and Teller have estimated the cross section for excitation of a nucleus.[6] The excitation by γ-rays is supposed to be a resonance absorption in which all the protons are set into oscillation together. The resonance character of the γ-ray excitation process was demonstrated by Baldwin and Klaiber[7] with 100-Mev bremsstrahlung.

According to the Goldhaber-Teller theory, the energy of the resonance is proportional to $A^{-1/6}$. The theory also predicts that $\int\sigma dE\propto A$, the atomic weight. If the bremsstrahlung spectrum from the betatron contained the same number of photons at all frequencies up to the maximum, and if the maximum energy of the betatron exceeded the energies at which the excitation of the nuclei occurred, then the photo-neutron yields observed by an experiment such as ours would be expected to be proportional to $\int\sigma dE$. A rough test of the theory can be made by allowing for the real shape of the spectrum and by allowing for the fact that the upper energy limit of the bremsstrahlung spectrum is not high enough to include all of the resonance curves for all the nuclei tested. Since the *intensity* spectrum from the betatron decreases approximately linearly by a factor of three between zero energy photons and maximum energy photons, the *number* spectrum is

$$N\propto 3(E/h\nu)-2.$$

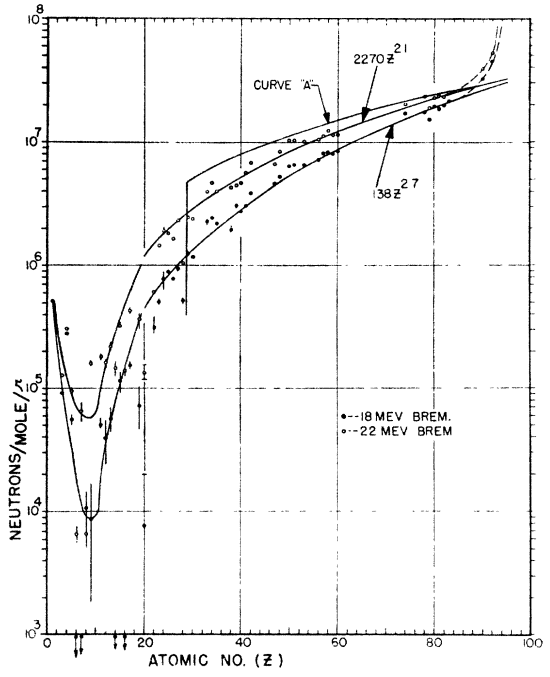

FIG. 4. Yields of photo-neutron sources for two bremsstrahlung spectra. Probable errors are shown by vertical lines through each point. Curve A is the yield predicted by Goldhaber and Teller which has been corrected for the 22-Mev bremsstrahlung spectrum and normalized at $Z=90$. The observed yield at $Z=29$ is about half the predicted yield because approximately half of the resonance is below 22 Mev.

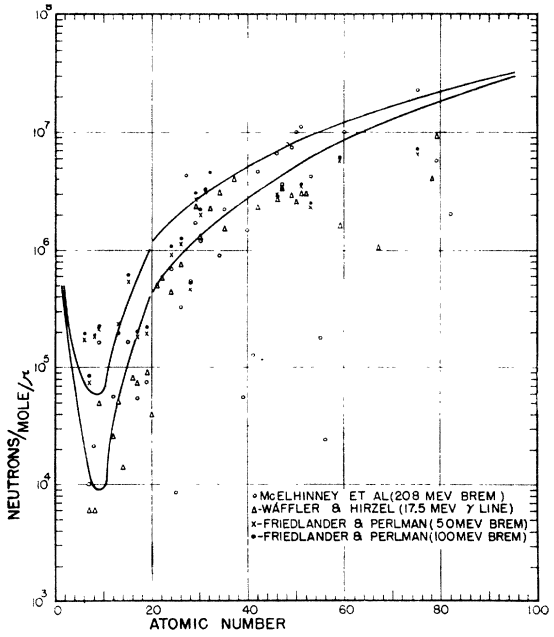

FIG. 5. Comparison of photo-neutron yields. Solid curves indicate results by Kerst and Price. The yields measured by McElhinney *et al.* are absolute whereas those measured by Wäffler and Hirzel, and Friedlander and Perlman are relative and have been normalized at $Z=47$.

[6] M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).

[7] G. C. Baldwin and G. C. Klaiber, Phys. Rev. **71**, 3 (1947); Phys. Rev. **73**, 1156 (1948). See also McElhinney, Hanson, Becker, Duffield and Diven, Phys. Rev. **75**, 552 (1949).

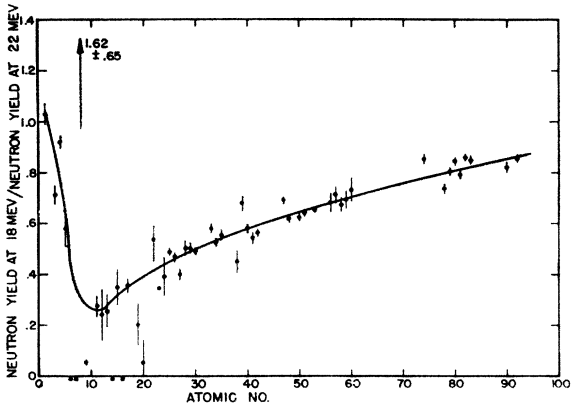

FIG. 6. Ratio of photo-neutron yields with 18-Mev and 22-Mev bremsstrahlung spectra.

For the sake of simplicity we now assume that the resonance is narrow. Then the yield $Y \propto N_{RES} \int \sigma dE \propto N_{RES} A$ but $N_{RES} \propto 3(A/A_0)^{1/6} - 2$, where A_0 is the atomic number for which the resonant energy, $h\nu_{RES}$, is equal to the upper limit of the bremsstrahlung spectrum, E. Then $Y \propto [3(A/A_0)^{1/6} - 2]A$.

Figure 4, curve A, shows this calculated yield with $Z_0 = 29$ for 22 Mev as is observed.[7] This curve has been made coincident with the extrapolated yield at $Z = 90$ assuming that all of the thorium resonance curve is below 22 Mev. One sees that at $Z = 29$, for copper, the calculated yield is 1.75 times the observed yield. This is approximately what is expected if half of the resonance curve for copper exceeds the upper limit of the bremsstrahlung spectrum. Thus to the rough approximation of these estimates the Goldhaber-Teller theory is in agreement with the observed yield. Levinger and Bethe[8] find $\int \sigma dE \propto ZN/A$. This gives approximately the same curve A.

It has not been possible to fit the yield curve by assuming Breit-Wigner resonance curves of widths 0 to 8 Mev and by assuming the threshold energy to be one-half of the resonance energy. The integrated cross section was kept proportional to A. The results follow the narrow resonance curve except in the vicinity $Z = 29$. Although the energy dependence of our neutron detector is not tested, it ought to be similar to the dependence for the "long counter."[9]

The ratio of yield for 18 Mev to the yield for 22 Mev, Fig. 6, shows that the resonance curves must have gradually varying shapes as a function of Z, because (above $Z = 20$) the ratio changes slowly with atomic number. Below $Z = 20$ the yields, and consequently the ratio of yields is strongly influenced by high thresholds.

Nickel yields for 18 Mev and for 22-Mev bremsstrahlung are each about 45 percent of the general trend. Chemical analysis shows 98.45 percent Ni, 0.86 percent Fe and traces of Sn and Sb. Thus impurities cannot be responsible for the low yields. The fact that the ratio for nickel (Fig. 6) is nearly the same as the ratio for neighboring elements suggests that the shape of the cross-section curve is not radically different for nickel, but it is possible to have a normal ratio with low yields as a result of high neutron binding energy and unusually large (γ,γ) or (γ,p) competition. It would be valuable to know the thresholds and the shapes of cross-section curves for nickel.

[8] J. S. Levinger and H. A. Bethe, Phys. Rev. **77**, 756 (1950).
[9] A. O. Hanson and J. L. McKibben, Phys. Rev. **72**, 673 (1947).

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Correlation of Geiger Counter and Hall Effect Measurements in Alloys Containing Germanium and Radioactive Antimony 124

G. L. PEARSON, J. D. STRUTHERS, AND H. C. THEUERER
Bell Telephone Laboratories, Murray Hill, New Jersey
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The distribution of the solute atoms in ingots of germanium antimony alloys which were prepared according to a prescribed cooling cycle have been measured. The solute was Sb^{124} and its distribution was measured with a Geiger counter. In addition, Hall measurements were made which show that in the impurity saturation range each antimony atom contributes one conduction electron. This makes it possible to determine the solute distribution in non-radioactive samples by the Hall method.

I. INTRODUCTION

THE electric properties of germanium containing small amounts of antimony depend on the concentration of antimony present. In preparing the ingots they are slowly cooled to control the antimony separation which occurs on solidifying a binary alloy. Because of the small concentrations of the antimony, 10^{-2} to 10^{-6} percent by weight, it is impossible to determine quantitatively its distribution in the ingot by ordinary chemical or spectrochemical methods. The availability of a radioactive isotope of antimony (Sb^{124}) offers a solution to this problem.[1]

[1] A preliminary account of this work was presented by the authors at the American Physical Society meeting in Chicago (November, 1948); Phys. Rev. **75**, 344 (1949).