that the test itself is a simple precise way of determining the nature of these doublets.

It is a pleasure to acknowledge the cooperation of the Radiation Laboratory of the University of California, and particularly of G. Bernard Rossi and the crew of the 60-in. cyclotron, in securing the many bombardments essential in this work. We are also grateful to the University of Chicago and the betatron operating crew for a bombardment received from them. Finally, the help of Dr. D. M. Van Patter and Dr. A. E. S. Green in the preparation of this paper is gratefully acknowledged.

PHYSICAL REVIEW VOLUME 94, NUMBER 6 JUNE 15, 1954

Systematics of Photoproton Reactions*

E. V. Weinstock and J. Halpern
University of Pennsylvania, Philadelphia, Pennsylvania
(Received March 3, 1954)

The photoproton yields from the elements Ta, Pt, Pb, W, and Au have been determined for betatron bremsstrahlung bombardment at 22-Mev peak energy by the use of zinc sulfide detectors and a 40-μsec betatron pulse duration. The yields, as measured in photoprotons per mole per roentgen, are as follows: Ta, 5.7×10^4; Pt, 2.9×10^4; Pb, 5.8×10^4; W, 5.2×10^4; and Au, 1.9×10^4. These values, along with previous determinations, permit a study of general behavior of photoproton yields for all Z values throughout the periodic table. Comparisons with calculations based on the evaporation model show good agreement with experimental trends up to a Z of 50, after which the measured yields are too high by factors ranging from 10 to 10^4. Calculations based on the direct photoelectric process give better agreement.

INTRODUCTION

In a systematic study of the proton yields from elements bombarded with bremsstrahlung radiation of moderate energy, almost all previous investigations have been limited to elements of either low or medium atomic number.[1–5] With the exception of cerium and bismuth, reported by Toms and Stephens, the elements studied have been confined to the region below a Z of 50. The present paper describes the measurement of photoproton yields from the elements tantalum, tungsten, platinum, gold, and lead. With these additions the systematics of yields as a function of Z is fairly complete except for a gap between a Z of 50 and 73. It now becomes possible to explore the general trend of the results with the proposed mechanisms for photonuclear reactions.

The twenty elements previously reported by this laboratory[1] were studied with betatron bremsstrahlung of 23.5-Mev maximum energy. Diven and Almy used 20.8 Mev, Butler and Almy 22.5 Mev, and Toms and Stephens 22.5 Mev for magnesium and 24 Mev for indium, cerium, and bismuth. The present work employed 22-Mev bremsstrahlung and the photoprotons were directly detected, as previously, with the use of zinc sulfide scintillators.

APPARATUS AND PROCEDURE

The experimental apparatus is essentially the same as that described in detail in reference 1. Briefly, a strongly collimated bremsstrahlung beam of maximum energy 22 Mev strikes the target, consisting of a thin sheet of the element under investigation, placed at an angle of 60 degrees with respect to two identical ZnS scintillators diametrically opposite each other. The detectors are kept fixed at an angle of 90 degrees to the photon beam and subtend an angle of 10 degrees at the target. After passing through the target, the photon beam strikes a 2-g/cm^2 tantalum target contained in a neutron detection assembly, and the number of neutrons from the Ta(γ,n) reaction is used to monitor the bremsstrahlung intensity. Additional monitoring makes use of an ionization chamber and current integrator, which in turn are calibrated in terms of a Victoreen r thimble imbedded in 4 cm of Lucite.

The scintillation pulses from the ZnS screens are detected with the use of 5819 photomultiplier tubes feeding model-100 amplifiers with 1-μsec delay line clipping and into ten-channel integral pulse-height analyzers. For high-Z targets, the combination of low proton yield and large light particle scattering makes discrimination against light particle pileup critical, and with a one-microsecond betatron pulse duration the beam intensity must be reduced prohibitively to achieve sufficient discrimination for the proton pulses. It was this, in fact, which limited the previous study to elements with a Z below 50. By the use of tech-

* Supported in part by the U. S. Air Research and Development Command and the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.
[1] A. K. Mann and J. Halpern, Phys. Rev. **82**, 733 (1951).
[2] B. C. Diven and G. M. Almy, Phys. Rev. **80**, 407 (1950).
[3] W. A. Butler and G. M. Almy, Phys. Rev. **91**, 58 (1953).
[4] M. E. Toms and W. E. Stephens, Phys. Rev. **82**, 709 (1951).
[5] M. E. Toms and W. E. Stephens, Phys. Rev. **92**, 362 (1953).

TABLE I. Target characteristics and yield data.

Element	Z	A	Percent abundance	Target thickness (mg/cm²)	(γ,p) threshold (Mev)	Resonance peak energy	Measured yield (protons/mole/r)	Percent error	Predicted yield (evap.)
Tantalum	73	181	100	124	6.17	14.1	5.7×10^4	30	1.6×10^2
Tungsten	74			136		14.1	5.2×10^4	31	1.2×10^2
		180	0.14		6.62				
		182	26.2		7.08				
		183	14.3		7.30				
		184	30.7		7.52				
		186	28.7		7.96				
Platinum	78			157		13.9	2.9×10^4	51	40
		190	0.012		6.16				
		192	0.78		6.59				
		194	32.8		7.01				
		195	33.7		7.23				
		196	25.4		7.44				
		198	7.2		7.86				
Gold	79	197	100	112	5.72	13.8	1.9×10^4	83	29
Lead	82			140		13.7	5.8×10^4	30	13
		204	13		6.53				
		206	26		6.93				
		207	21		7.12				
		208	52		7.33				

niques similar to those reported by Keegan,[6] the betatron pulse duration was lengthened to 40 microseconds for the work reported herein, whereupon discrimination against pileup was unambiguous as indicated by the integral bias curve for tantalum shown in Fig. 1. Also included in the figure are a curve representing the background obtained without a target in the scattering chamber, and the net curve with background subtracted. These data were taken with a photon beam intensity of 5.9 roentgen per minute at the target, whereupon the net useful proton counting rate was 0.2 counts per minute for a tantalum target of 124-mg/cm² thickness, as obtained by extrapolation of the net curve to zero bias.

Targets were in the form of foils of commercial purity and natural isotopic composition. The low proton yield required the use of thick targets as listed in Table I, which necessitated a large target absorption correction calculated as outlined in reference 1. The calculated corrections increased the observed yield values by a factor of 2.5.

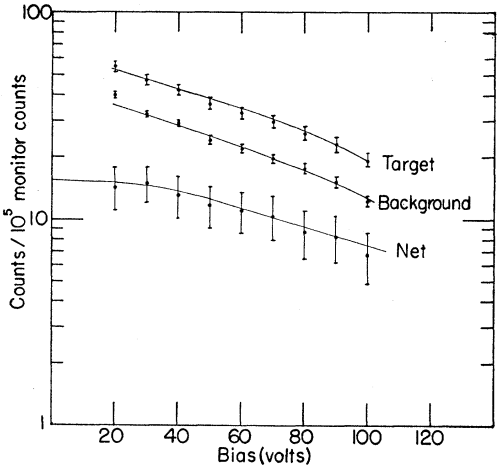

FIG. 1. Integral bias curves for a 124-mg/cm² tantalum target. Net count is obtained by subtracting background curve from target curve and extrapolating to zero bias.

Since measurements were made only at one angle, total yields were computed from the known geometry of the apparatus assuming spherical symmetry for the proton angular distributions. Although angular distribution data for heavy elements are sparse,[5,7,8] it is not believed that errors greater than 25 percent can enter as a result of the assumption of isotropy.

The possibility existed that some or all of the protons observed might arise from (n,p) reactions induced in the target by fast neutrons in the photon beam or present as background at the target position. Consequently it was deemed necessary to measure the fast neutron flux intercepted by the targets making use of the Lucite-zinc-sulfide fast neutron detector developed by Hornyak.[9] By further expansion of the betatron pulse duration to 100 microseconds and reduction of the photon beam intensity by a factor of 3000, it was possible to place the neutron detector in the direct betatron beam without excessive pileup. By calibration of the neutron detector efficiency using a radium-beryllium neutron source of known strength, and using reported (n,p) cross sections,[10] it was possible to show that any contribution to the measured proton yields from fast neutron reactions could not exceed a few percent at the most.

[6] T. J. Keegan, Rev. Sci. Instr. **24**, 472 (1953).

[7] Mann, Halpern, and Rothman, Phys. Rev. **87**, 146 (1952).
[8] M. M. Hoffman and A. G. W. Cameron, Phys. Rev. **92**, 1184 (1953).
[9] W. F. Hornyak, Rev. Sci. Instr. **23**, 264 (1952).
[10] B. L. Cohen, Phys. Rev. **81**, 184 (1951).

RESULTS AND DISCUSSION

The corrected absolute yield values are listed in Table I and plotted against Z in Fig. 2. The errors shown are those due to counting statistics as a result of the low yields inherent in heavy element photoproton emission. Also shown in the figure are all previous yield measurements made under comparable conditions. Included are the yields of Mann and Halpern taken using the method of this paper, and the photographically detected photoproton yields of Butler and Almy (B & A), Diven and Almy (D & A), and Toms and Stephens (T & S).

In a comparison of the data with the theories for photonuclear reactions we will be concerned with the general trends of the yields as opposed to individual fluctuations of neighboring elements. With increasing Z, the yields rise to a maximum at nickel ($Z=28$) and then drop rapidly until a Z of 50, whereupon they show a much slower drop at high Z values. An attempt will now be made to understand these trends on the basis of the evaporation model of nuclear reactions with corrections for the direct photoelectric ejection of a proton in the region of high Z, where this mechanism becomes relatively more important.

Following Weisskopf,[11] the (γ,p) cross section is:

$$\sigma(\gamma,p)=\sigma(\gamma)G_p. \tag{1}$$

Here $\sigma(\gamma)$ is the γ-ray absorption cross section and G_p the branching ratio, given by

$$G_p=F_p/\Sigma_b F_b, \tag{2}$$

where the summation is extended over all particles

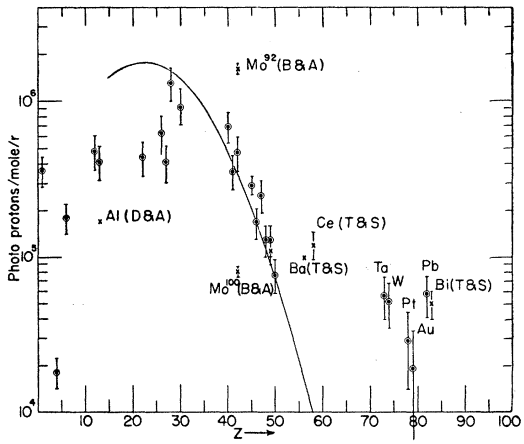

FIG. 2. Summary of published photoproton yield data. Unlabeled points are those of Mann and Halpern; crosses are data from Diven and Almy (D & A), Butler and Almy (B & A), and Toms and Stephens (T & S); points labeled only with chemical symbols are those obtained in the present work. The solid curve is the yield *vs* Z as predicted by the calculations described in the text.

[11] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), pp. 340–374.

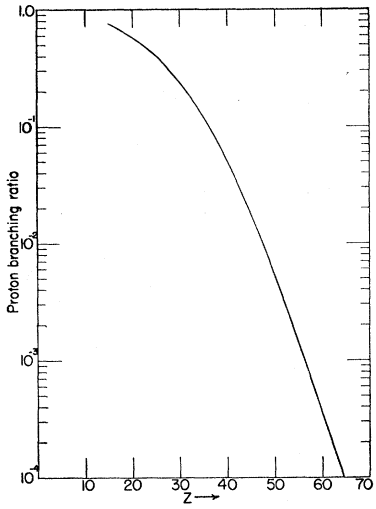

FIG. 3 Proton branching ratio as a function of Z.

that can be emitted in the reaction, and

$$F_b=\frac{2M_b}{\hbar^2}\int_0^{\epsilon_{\max}}\epsilon\sigma_{cb}(\epsilon)w(\epsilon_{\max}-\epsilon)d\epsilon. \tag{3}$$

M_b is the mass of the emitted particle, ϵ its energy upon emission, and $\epsilon_{\max}$ the maximum value that ϵ can assume (given in the present case by $\hbar w-S_p$, where S_p is the energy with which particle b is bound to the compound nucleus); σ_{cb} is the cross section for formation of the compound nucleus by particle b, and w is the density of energy levels of the residual nucleus.

The photoproton yield per mole per roentgen is then

$$\begin{aligned}Y_p&=a\int_{E_{th}}^{E_\beta}\sigma(\gamma,p)N(E_\gamma,E_\beta)dE_\gamma\\&=a\int_{E_{th}}^{E_\beta}\sigma(\gamma)\frac{F_p}{\Sigma_b F_b}N(E_\gamma,E_\beta)dE_\gamma,\end{aligned} \tag{4}$$

where $N(E_\gamma,E_\beta)$ is the bremsstrahlung distribution at the energy E_γ when the betatron energy is E_β; E_{th} is the threshold for the reaction, and a represents Avogadro's number.

A calculation of the proton yields therefore requires a knowledge of the energy dependence of the total absorption cross section for each element; and data for the various quantities that are contained in the F's, i.e., the particle binding energies, and capture cross sections and the level densities. Sufficient approximations to these quantities are available so that one can compute the order of magnitude of proton yields to be expected for the various Z values.

In computing the branching ratios, the assumption was first made that particle emission other than proton and neutron could be neglected. Except in the special case of copper, where a sizable deuteron emission has

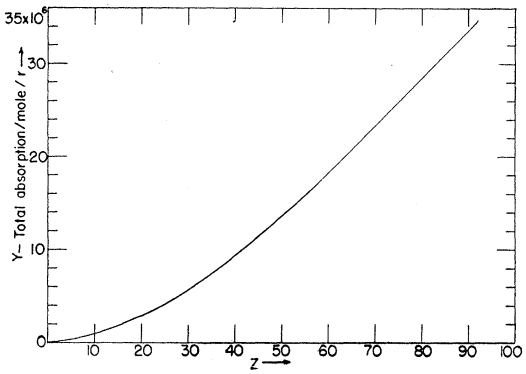

FIG. 4. Absorption yield as a function of Z.

been reported,[12] both barrier considerations and experiment rule out deuteron, triton, and alpha particle competition. Thus the branching ratio becomes simply $G_p=F_p/(F_p+F_n)$. Evaluation of the F functions involves ϵ_{max}, and thus the particle binding energies as well as the energies at which the photons are absorbed. Binding energies were computed from the mass formula,[13] and smooth curves were drawn through the values plotted as a function of Z. Since the photon absorption exhibits a resonance behavior, the peak position of the resonance as found from the systematic study of photoneutron reactions[14,15] was used for the photon energies. This approximation removed the branching ratios from the integral in Eq. (4) for the proton yield values.

F values have been plotted by Feld *et al.*,[16] with the assumption of exponential level densities for the residual nuclei. There is a further dependence on the nuclear radius which was chosen to be $1.3\times10^{-13}A^{\frac{1}{3}}$ cm for these evaluations. The resulting plot of the branching ratios as a function of Z is shown in Fig. 3.

There remains, then, the evaluation of the integral of the product of the absorption cross section and the bremsstrahlung distribution. The assumption of a narrow absorption resonance was again made through utilization of the bremsstrahlung intensity at the peak of the cross section *vs* energy curves. For the integrated cross section, the values predicted by Levinger and Bethe[17] from the dipole sum rules were inserted. The fraction of exchange in the neutron-proton interaction was assumed to be 0.5. The resultant absorption per mole per roentgen is plotted in Fig. 4 as a function of Z.

Since the absorption per mole per roentgen is an increasing function of Z and the branching ratio a decreasing function, the product of the two (representing the proton yield per mole per roentgen) will exhibit a peak as shown by the solid curve drawn through the measured values of Fig. 2. The peak occurs at $Z=23$, and between a Z of 28 and 50 the experimental points are in excellent agreement in absolute magnitude.

Even for the region below a Z of 28, the discrepancies with the above rough calculations is not serious especially in view of the fact that the level densities should depart radically from those assumed, the binding energies show large fluctuations, and it is known that the proton and neutron integrated cross sections up to 22 Mev do not add up to the predictions of the sum rules.[15]

The discrepancies, however, in the high-Z region are more serious as illustrated by the measured and predicted values listed in Table I. Observed yields are higher than those calculated by factors which get as large as three orders of magnitude, the agreement becoming progressively worse with increasing Z. Thus, even on the basis of such rough calculations, it is safe to say that the evaporation model as usually formulated is inadequate to explain the photoproton yields for elements of high atomic number.

As shown by Toms and Stephens[5] for the element bismuth, a considerable portion of the photoproton yield can be accounted for on the basis of the direct photoelectric emission of a proton without the formation of a compound nucleus. Using the formulation of Courant,[18] they found that the observed yields were within a factor of four of those predicted by this mechanism. We have performed a similar computation for tantalum and find the observed yields too high by a factor of ten. This is not serious, for as pointed out by Courant, a factor of 10 could easily be found in the calculations by modifying the assumptions regarding particle correlations in the nucleus and the shape of the nuclear potential. There is also a substantial quadrupole yield from the heavy elements,[19] and this could modify the proton energy distributions sufficiently to enhance the direct yield.

[12] P. R. Byerly and W. E. Stephens, Phys. Rev. **83**, 54 (1951).
[13] N. Metropolis and G. Reitwiesner, "Table of Atomic Masses," Los Alamos Scientific Laboratory Report NP-1980, 1950 (unpublished).
[14] Montalbetti, Katz, and Goldemberg, Phys. Rev. **91**, 659 (1953).
[15] R. Nathans and J. Halpern, Phys. Rev. **93**, 437 (1954).
[16] Feld, Feshbach, Goldberger, and Weisskopf, U. S. Atomic Energy Commission Report NYO-632 (unpublished).
[17] J. S. Levinger and H. A. Bethe, Phys. Rev. **78**, 115 (1950).
[18] E. D. Courant, Phys. Rev. **82**, 703 (1951).
[19] Halpern, Nathans, and Mann, Phys. Rev. **88**, 679 (1952).