Millisecond Gamma Emitter from Protons on Tantalum*

SHELDON D. SOFTKY†
Radiation Laboratory, Department of Physics, University of California, Berkeley, California
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A search for gamma emitters in the millisecond range of half-life was conducted, using the 32-Mev Berkeley proton linear accelerator. Foil targets of eighteen common elements were bombarded, and the activity made in them was followed between beam pulses with a gated scintillation counter. The only activity found of measurable yield was from Ta, and it exhibited a half-life of 5.5±0.3 milliseconds. Using a gated nine-channel pulse-height analyzer with the NaI(Tl) crystal counter, the excitation curve of this activity was measured and its gamma-ray spectrum deduced. Gamma rays of 0.35 Mev and 0.22 Mev are indicated by the pulse spectrum, and no β's are present commensurate with this half-life. Comparison of the excitation curve for this activity with those of Ta^{180} (8-hour) and W^{179} (30-min) leads to the tentative assignment of this decay to W^{180m}.

INTRODUCTION

SINCE the discovery of nuclear isomerism by Hahn[1] in 1921, over a hundred examples of it have been found among the known radioactive nuclei. In spite of powerful methods of electronic detection,[2] however, no isomers with half-life in the millisecond range have been discovered until very recently,[3] and yet isomeric half-lives have been found as long as years and as short as 10^{-10} second. Goldhaber and Sunyar[4] suggested that the absence of any half-lives of the millisecond magnitude might actually be equivalent to their improbability on an energy basis. On the basis of their semi-empirical classification of isomers, they found that the only combinations of spin change and gamma energy that could correspond to millisecond half-lives were E_γ about 50 kev for $\Delta I=2$, and E_γ about 800 kev for $\Delta I=3$. Since the variation of half-life with energy is a power law, these possible energies fairly sharply define the half-life and make the rarity of isomers with these decay energies simply equivalent to a rarity of millisecond half-lives.

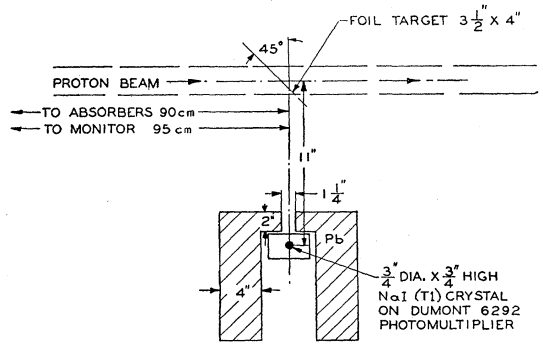

FIG. 1. Bombardment arrangement.

A way of searching for these activities that had not been extensively used until the last few years was made available by pulsed heavy-particle accelerators. This method consists of bombarding a target that is near a counter and gating the counter so that it can count only during the time between beam pulses. Providing the target did not get too active with longer-lived activities, one could detect by this means any activity whose half-life was of the order of the time between beam pulses or much shorter than this. This technique was successfully used by Alvarez[5] to find and identify the twelve-millisecond N^{12} which emits a very energetic beta, but had not been used to find gamma emitters, perhaps because only in the last few years have high-efficiency, short-dead-time gamma counters been available.

If, now, an activity is detected which exhibits a very short half-life and yet emits only gamma radiation or low-energy electrons which could be conversion electrons or photoelectrons, this activity must almost surely be a nuclear isomer since a K capture or beta emitter would have to be of very high energy indeed in order to have a half-life in the millisecond region. For example, the 27-msec B^{12} emits a β^- of 13.4 Mev and the 12-msec N^{12} emits a β^- of 16.6 Mev; a K capture among even the heaviest of elements could hardly be expected to occur with energies of this magnitude without appreciable positron emission.

The Berkeley 32-Mev proton linear accelerator is a pulsed machine which delivers 300-μsec long pulses fifteen times per second. This makes it almost ideal to use for the production of short-lived activities, and so a search was undertaken for gamma-ray emitters with half-lives in the range between one millisecond and 50 milliseconds which could be made by 32-Mev protons incident on a number of different elements.

METHOD

The geometry used for bombardment of the various targets is shown in Fig. 1. The counter consisted of a ¾-inch diameter by ¾-inch high NaI(Tl) crystal pack-

* This work was done under the auspices of the U. S. Atomic Energy Commission.
† Now at Tracerlab, Inc., Western Division, Richmond, California.
[1] E. L. Hahn, Chem. Ber. 54, 1131 (1921).
[2] S. DeBenedetti and F. K. McGowan, Phys. Rev. 74, 736 (1948).
[3] H. B. Mathur and E. K. Hyde, Phys. Rev. 98, 79 (1955).
[4] M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).
[5] L. Alvarez, Phys. Rev. 75, 1815 (1949).

aged with MgO around three sides and optically glued to the window of a DuMont 6292 photomultiplier tube. The photomultiplier was protected from stray magnetic fields by a Mu metal shield, and its gain was calibrated against the gamma ray of 0.662 Mev from Cs^{137}. The gain of the entire system made this gamma ray give a pulse height for photo-absorption of about 50 to 70 volts. The pulse spectrum due to Cs^{137} is shown dotted in Fig. 2, the strong peak being the photo-absorption line.

An electronic gating unit was built which delivered a total of six gate pulses in tandem which followed the end of the beam pulse by six milliseconds. These gate pulses were fed separately to each of six scalers so that the only time a scaler could accept input pulses from

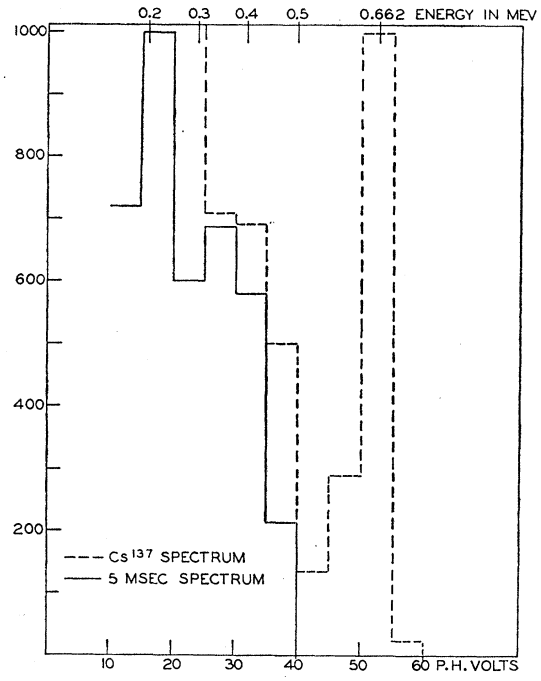

FIG. 2. Pulse-height spectra of Cs^{137} and 5.5-msec activity.

the scintillation counter was within the duration of the gate. Figure 3 shows the electronic arrangement for this counting experiment. The scaler discriminators were set at the same pulse height, about that of a 100-kev gamma ray, and checked frequently. The gates on the scalers were equal in width so that the counting rates on the scalers formed a decay curve for the activity after background was subtracted away. Background consisted of two counting rates: the first was simply that due to the beam alone with no target in place; the second was due to the long-lived activities made in the target and was measured by counting for the same length of time as an actual measurement, but with no beam and beginning 30 seconds after the beam had been turned off.

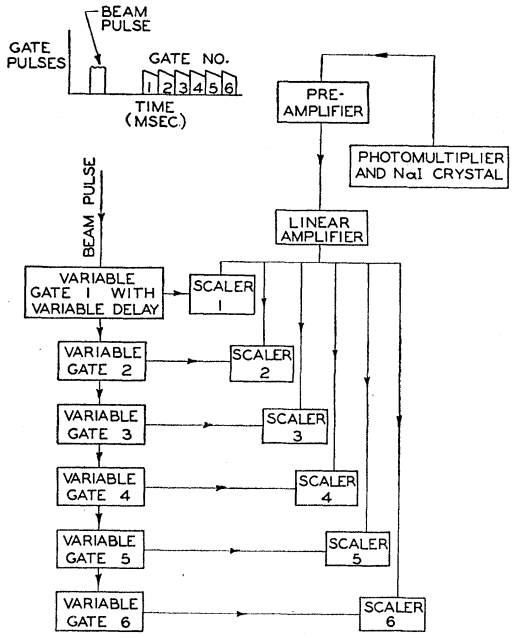

FIG. 3. Block diagram of electronics.

Foils approximately 10^{21} atoms/cm^2 thick of Pb, Pt, W, Mg, Al, Ni, C, Cu, Zn, Mo, Ag, Cd, In, Sn, Ta, Bi, Be, and U were bombarded in the manner described, and the only activity observed that indicated a short-lived decay was from Ta as a target. The gate widths and delay were such that an abundant activity of half-life from about 2 msec to 25 msec would probably have been detected. Thick targets of W, Ta, and Pb were bombarded, and the ratio of activities of the Ta to the W and Pb targets was greater than 50:1. This ratio was obtained by monitoring the beam with an argon-filled ion chamber during the runs on different targets. The incident beam energy was varied with Al absorbers, and the yield and half-life for twelve energies down to 14 Mev were measured for the thick Ta target. This gave ten good measurements of the half-life, the average of which is plotted in Fig. 4. The gates were 2.5 msec wide for these runs and began 5 msec after the end of the beam pulse. The total spread of these ten measurements of half-life is less than five percent, and the average value is 5.5 ± 0.3 msec for the half-life, the error based on the extreme decay curves which can be drawn through the six points in Fig. 4.

In order to verify that the activity found was indeed an isomer, its gamma-ray spectrum was investigated and a thin target excitation function obtained. For this purpose a nine-channel pulse-height analyzer developed by Bowman, Thomas, and Gantz[6] was used. A beam monitor devised by Kitchen,[7] consisting of a DuMont

[6] H. R. Bowman and R. E. Thomas, University of California Radiation Laboratory Report UCRL-2164 (unpublished).
[7] Sumner Kitchen, University of California Radiation Laboratory Report UCRL-2495 (unpublished).

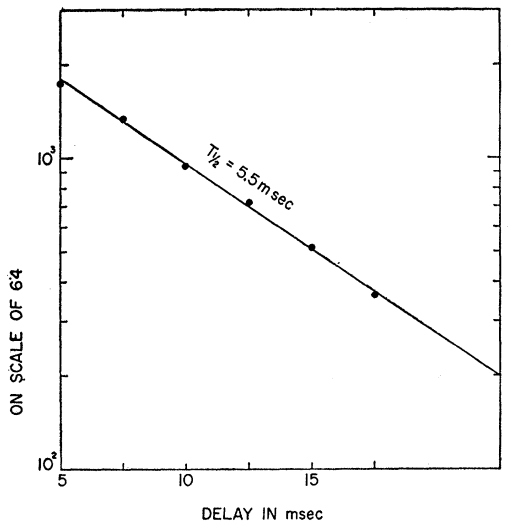

FIG. 4. Decay curve of 5-msec activity.

6292 photomultiplier looking at air ionized by the beam, was used since this type of monitor is linear with beam intensity whereas an ion chamber is not. The ion chamber is not linear because the highly focused beam of the linear accelerator "uses up" all the available atoms of gas in the ion chamber by ionizing them, so that further increases in beam intensity do not give equal increases in ion current.

With the pulse-height analyzer gated so that it accepted pulses only during the time interval from 7 msec after the beam till 13 msec after the beam, Ta foils 0.004 inch thick were bombarded so that the beam passed through the target at 45°, as shown in Fig 1; foils were changed frequently to keep background of long-lived activities down. Use of a pulse-height analyzer actually made the measurements from thin targets possible, since most of the total background was due to either very small or very large pulses. The pulse spectrum measured is shown as the solid histogram of Fig. 2. It is the average spectrum of five runs at different proton energies, and the separate spectra measured at these energies show that the gamma-ray spectrum does not change with proton energy. Even though the total background due to both causes mentioned changed both in spectrum and in magnitude, averaging 20 to 30 percent, the net spectrum did not. That no beta rays were present in the activity was shown by the fact that the pulse spectrum from the NaI crystal consisted mostly of relatively low-energy pulses. If betas were present with this half-life, they would have to be well above 5 Mev in energy, and betas this energetic traversing a crystal this large would have caused very large pulses. Also the pulse spectrum from the crystal, were it due to betas, would certainly have been changed in shape by the introduction of one inch of Be between target and crystal, and it was not. The effect of the Be was simply to smear the spectrum slightly and to attenuate it as much as would be estimated for gamma rays of this energy.

The excitation curve consists of the total net yield measured at seven different proton energies down to threshold, which is between 13 and 14 Mev. The beam energy was changed by Al absorbers and was calculated according to the curves of Aron *et al.*[8] for the middle of the Ta target. It should be pointed out that it was necessary to have a long air path to the target in order to keep activity from the Al absorbers from raising background, and since the beam was scattered somewhat by the absorbers, undoubtedly for the thicker absorbers some of the monitored beam never reached the target. This means that the measured excitation curve should be multiplied by some correction factor which monotonically increases for decrease in proton energy. The effect of this correction would be to make the true curve rise less rapidly from threshold than the measured one, to make the true peak at about 22 Mev more marked than the measured one, and to make the threshold as obtained by extrapolation lower than if obtained from the measured curve. Since this correction would not markedly change any conclusions drawn from the observed curve, it has not been made, and the observed curve is shown in Fig. 5.

In an attempt to identify the 5.5-msec activity, thin-target excitation curves were obtained for the 8-hour Ta^{180} and the 30-minute W^{179} from protons on Ta. Stacks of 0.002-inch Ta foils were bombarded with 31-Mev protons and the activities in each foil followed with the scintillation counter biased low enough to detect K x-rays from the decay. The composite decay curves were resolved for these two half-lives, and exhibited no activities in the range of 2 minutes to 30 minutes, nor measurable amounts of activities longer than 8 hours. This means that the 5-min activity assigned to W^{179} and listed in reference 9 probably does not exist, since it was supposedly made by 20-Mev protons on Ta.

A range-energy curve for Ta was obtained by interpolating between those for Ag and Pb given in Aron *et al.*,[8] e.g., by assuming that

$$\frac{R_{\mathrm{Ag}}(E_1)}{R_{\mathrm{Pb}}(E_1)}=K_1\frac{(Z/A)_{\mathrm{Pb}}}{(Z/A)_{\mathrm{Ag}}},$$

and using this to get a K_2 also for E_2. For $E_1=32$ Mev, $K_1=0.92$; for $E_2=13$ Mev, $K_2=0.88$. Then, with $K=0.90$, it was assumed that

$$\frac{R_{\mathrm{Ta}}(E)}{R_{\mathrm{Pb}}(E)}=K\frac{(Z/A)_{\mathrm{Pb}}}{(Z/A)_{\mathrm{Ta}}}.$$

The resulting excitation curves for Ta^{180} and W^{179} are plotted in Fig. 5 on the same energy scale as the 5.5-msec activity.

[8] Aron, Hoffman, and Williams, U. S. Atomic Energy Commission Report AECU-663, 1949 (unpublished).

DISCUSSION OF RESULTS

In accordance with the properties of a nuclear isomer, the above experiments show the existence of an isomer which has a half-life of 5.5 msec and a threshold for formation from protons on Ta of about 13 Mev. That it is an isomer is shown by the fact that it emits no betas of energy appropriate to this short half-life, and also that all the mass numbers to which it could possibly be assigned already either have activities of other half-life assigned to them or are stable. Table I shows the Segrè Chart for this region of Z and A.

That this activity is either Ta^{180} or W^{179} is ruled out because its excitation curve is noticeably different from the curves of those activities, and in accord with the mechanism for formation of isomers, it would have to be the same to be isomeric with either of them; i.e., isomers result from the falling down into a metastable state of a residual nucleus of high excitation, and it is this state for which we are actually measuring the excitation curve. That the activity is Ta^{179} is ruled out because its threshold would have to be about 8 Mev higher than that of Ta^{180}, which already has too high a threshold. Two assignments which cannot be ruled out on the basis of threshold and shape of the excitation curve are W^{181} and W^{180}. These would be possible by $Ta^{181}(p,n)W^{181}$ and $Ta^{181}(p,2n)W^{180}$, respectively. The observed threshold corresponds well with the Coulomb barrier height $Z/A^{\frac{1}{3}}$ (Mev) for protons penetrating the Ta nucleus, and the peaked shape of the excitation curve is what one might expect from a (p,xn) reaction. Of these two, $Ta^{181}(p,2n)W^{180}$ seems more likely because it is peaked where one would expect it to be relative to the measured $Ta^{181}(p,3n)W^{179}$ and to other known $(p,2n)$ reactions. All other nuclides except Ta^{181} itself

TABLE I. Isotope chart for region of Ta.

Hf $Z=72$	Ta $Z=73$	W $Z=74$
Hf^{180}	Ta^{181}	W^{182}
5.5 hr	22 μsec	
γ's of 0.057	γ's of 0.345	Stable
0.093	0.136	26.3%
0.214	0.481	
Stable	Stable	
35.4%	100%	
Hf^{179}	Ta^{180}	W^{181}
19 sec	8 hr	140 days
γ 0.15	β^- 0.6	γ 1.83
	γ 0.09	e^- 0.07
Stable	x-rays	x-rays
13.8%		(Threshold ~13 Mev)
Hf^{178}	Ta^{179}	W^{180}
	600 days	Stable
Stable	γ 0.7	0.126%
27.1%	e^- 0.01	
		(Threshold ~13 Mev)
Hf^{177}	Ta^{178}	W^{179}
	9 min	30 min
Stable	β^+ 1.06 3%	No γ
18.4%	γ 1.5 3%	x-rays
	e^- 0.08 50%	
	x-rays	

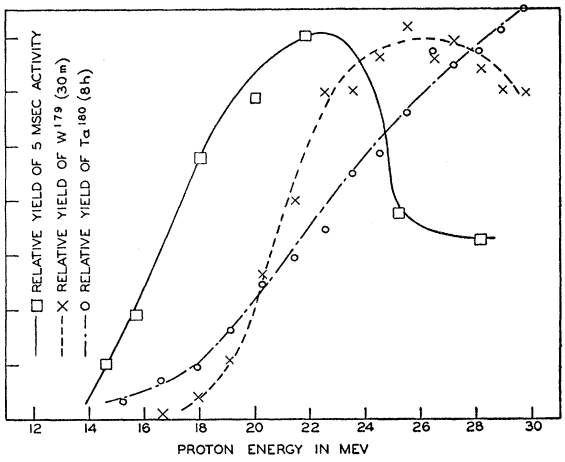

FIG. 5. Excitation functions for three activities from Ta+p.

can be ruled out on the basis that the energetic threshold plus the Coulomb barrier to protons within the compound nucleus add up to a total threshold that is too high to fit the curves obtained. By this means all such reactions as $(p,2pn)$ or (p,pxn) are ruled out. The assignment of the activity to W^{180} is also consistent with the fact that it was not observed from protons on W, since to make it from W^{182} would require a $(p,p2n)$ reaction that is energetically of too high a threshold to see, and W^{180} (stable) is only 0.13 percent abundant, so the (p,p') reaction would make little of it. Perhaps a further experiment could be done on separated or enriched W^{180} to verify the assignment. Nuclear isomers of even Z and A are rather rare, there being five in the newest isotope table available.[9] These are all in this region of $Z=72$, being Hf^{180m}, Os^{186m}, Pb^{202m}, and two of Pb^{204m}. They are unlikely simply on the basis of having both neutrons and protons all paired, but the assignment of the 5.5-msec activity to an excited state of stable W^{180} is not particularly difficult to reconcile with this scarcity, since those which are known are scattered over this very region of Z and A.

From the relatively crude pulse-height spectrum of Fig. 2, it can only be inferred that there is a gamma ray of about 0.35 Mev in the decay of this activity, and very probably one of about 0.22 Mev corresponding to the lower peak. There could easily be lower-energy gammas present that were not counted. Goldhaber and Sunyar[4] predicted on the basis of their semiempirical classification of isomers that the only possibilities for an isomer to have a half-life in the millisecond range would correspond either to a spin change of two for which $E_\gamma \simeq 50$ kev, or spin change of three, for which $E_\gamma \simeq 800$ kev. They also show a distribution in spin and parity for the first excited states of even-even nuclei, which overwhelmingly favors $I=2, +$. On the basis of these facts the most comforting guess one can make about the

[9] Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953).

5.5-msec transition is that it is $\Delta I=2$, no change in parity, which would make it electric quadrupole. Then one would have to hypothesize a low-energy gamma in the spectrum, which was the half-life-determining transition.

CONCLUSIONS AND SUMMARY

From the bombardment of several common elements with 32-Mev protons, only one gamma emitter with half-life between 2 msec and 25 msec was found. On the basis of thresholds for neighboring activities and shapes of their excitation curves and that of the new activity, this 5.5-msec half-life activity from $p+$Ta has been tentatively assigned as a metastable state of stable W^{180}. No sure assignment can be made to the transition concerning spin and parity.

ACKNOWLEDGMENTS

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Neutron Spectrum and Absolute Yield of a Plutonium-Beryllium Source*

LEONA STEWART
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico
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A Pu-Be neutron source has the advantages of high neutron yield, low gamma-ray intensity, and very long half-life. The neutron spectrum from such a source, composed of 13 g of plutonium and 7 g of beryllium as $PuBe_{13}$ (density 3.7 g/cc), was measured by means of proton recoils in nuclear research emulsions. The data obtained from 2057 observations indicate the neutrons have a maximum energy of approximately 10.5 Mev with broad intensity maxima at 4.0, 7.2, and 9.7 Mev. The observed spectrum shows marked similarities with published data on neutrons from Po-Be and Ra-Be sources. A comparison is made between the present data and previously reported energy levels in the residual carbon nucleus. The absolute yield of neutrons above 0.5-Mev energy is 1.2×10^6 neutrons/sec.

I. INTRODUCTION

ALTHOUGH neutron sources employing the (α,n) reaction have been in use for over two decades, few precise spectral measurements have been reported. Of the early work,[1] probably the most widely known are the data of Richards and of Demers obtained by proton recoil measurements in photographic plates. Both these measurements were made with the old Ilford half-tone emulsions which were characterized by low silver halide concentration and consequently poor energy resolution. In addition, the energy calibration of these emulsions was not accurately known. It is not surprising therefore to find some disagreement in these early measurements.

More recently, Whitmore and Baker,[2] Guier, Bertini, and Roberts,[3] Gursky, Winnemore, and Cowan,[4] and Elliot, McGarry, and Faust[5] have used various methods[6] to obtain quite precise data on Po-Be sources by making use of the many refinements achieved during the past decade in neutron spectrum measurements. (For a discussion of these methods see Barschall, Rosen, Taschek, and Williams[7] and Rosen.[8])

The most accurate work appearing in the literature on Ra-Be was done by Hill,[9] who made ionization chamber measurements employing a coincidence-anticoincidence arrangement which permitted high statistical accuracy as well as good resolution.

Although Po-Be is presently more widely used as a neutron source because of its availability, Pu-Be possesses many advantages over Po-Be and may be expected to replace it where neutron intensity is not the determining factor. Po-Be is a mixture, and the spectrum therefore varies with grain size. Even after

* Work performed under the auspices of the U. S. Atomic Energy Commission. Submitted as a Master's thesis to the University of Texas, January, 1953.

[1] N. Feather, Proc. Roy. Soc. (London) **A142**, 3 (1933); P. Auger, J. phys. radium **4**, 749 (1933); J. R. Dunning, Phys. Rev. **45**, 586 (1934); M. Blau, J. phys. radium **5**, 61 (1934); T. W. Bonner and Mott-Smith, Phys. Rev. **46**, 258 (1934); G. Bernardini and D. Bocciarelli, Atti. accad. nazl. Lincei **24**, 132 (1936); G. Bernardini, *Kernphysik Vortrage*, Zurich, 1936 (unpublished); Perlman, Richards, and Speck, Atomic Energy Commission Report MDDC-39, 1943 (unpublished); H. T. Richards, Atomic Energy Commission Report MDDC-1504, 1944 (unpublished); P. Demers, Montreal Report MP-74 and MP-204, 1945 (unpublished).

[2] B. G. Whitmore and W. B. Baker, Phys. Rev. **78**, 799 (1950).

[3] Guier, Bertini, and Roberts, Phys. Rev. **85**, 426 (1952).

[4] Gursky, Winnemore, and Cowan, Phys. Rev. **91**, 209 (1953).

[5] Elliot, McGarry, and Faust, Phys. Rev. **93**, 1348 (1953).

[6] The first two measurements were made with photographic emulsions, the second with an ionization chamber, and the third with a coincidence spectrometer.

[7] Barschall, Rosen, Taschek, and Williams, Revs. Modern Phys. **24**, 1 (1952).

[8] L. Rosen, Nucleonics **11**, No. 7, 32 and No. 8, 38 (1953).

[9] D. L. Hill, Atomic Energy Commission Report AECD-1945, 1947 (unpublished).