the same as the corresponding one for the S-matrix. The regularized infinities will appear as some multiplicative constants and will not depend on the form of the wave function $\langle V|v(x_1,x_2)\rangle$. The terms like SE and MI"SL" on the other hand, seem to involve $\langle V|v(x_1,x_2)\rangle$ more deeply, the arguments of the latter being integration variables with respect to which the most dangerous integrations are being performed. The function $\langle V|v(x_1,x_2)\rangle$ certainly cannot be represented by plane waves, and by some particular choice of it the infinities can be even made to disappear. It seems therefore that some additional techniques have to be developed in order to regularize such terms for a wide enough class of wave functions. If on the other hand the S-matrix techniques were to turn out to be adequate for the above purposes, it would not be, in the author's opinion, without some adaptations made in the actual computation rules of the integrals concerned. In this case one might even hope to get a deeper insight into the physical meaning of these methods. In addition to those already given, Fig. 3 shows ten nets completing the interaction patterns in the e^4-approximation. These are less interesting, as they may be decomposed into simpler parts encountered before. It might, nevertheless, be of some interest to observe that the characteristic vacuum polarization loop "attached" to one photon line appears where the external potential would also appear, should it be postulated. The meaning of this loop is simply an (infinite) potential produced by the Dirac "sea," and it might be physically considered meaningless. With regard to all nets given above which are not already symmetric with respect to both particles, we finally observe that they are to be repeated with the roles of the particles exchanged.

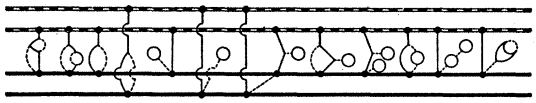

FIG. 3. Ten nets completing the interaction patterns in the e^4 approximation.

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Reactions of Cesium with 240-Mev Protons*

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Absolute yields of radionuclides produced in the spallation of cesium with 240-Mev protons have been measured from barium to ruthenium. The yields are based on the known cross section for the monitor reaction $Al^{27}(p,3pn)Na^{24}$. In agreement with the well-established pattern for high-energy spallation yields, the results indicate that the majority of reactions occur as primary knock-on collisions in which the incident proton interacts with only a few nucleons. This primary interaction is followed by a dissipation of the residual nuclear excitation, whose energy spread is very broad, leading to a distribution of products which reach far down the nuclide chart.

Measurement of absolute x-ray counting efficiencies for the K x-rays of Xe^{131} and Mn^{55} in a methane beta-proportional counter and in a helium-alcohol G-M tube is also reported.

I. INTRODUCTION

THE nuclei which result when target nuclei are struck by high energy projectiles have been investigated in several instances by measurement of the yields of various radioactive products.[1]

An outstanding feature of high-energy reactions is the wide distribution of spallation products which result from dissipation of the excitation energy of the struck nuclei. In all cases where spallation has been studied at energies above 100 Mev, it has been found that while the distribution of products spreads far down the periodic chart from the target element, the distribution of yields is sharply accentuated in the region close to the target. A proposal by Serber and enlarged upon by Goldberger[2] gives a qualitative mechanism whereby fairly small excitation energies may be transferred to the target nucleus by projectiles of very great energy.

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[1] Cunningham, Hopkins, Jr., Lindner, Miller, O'Connor, Perlman, Seaborg, and Thompson, Phys. Rev. **72**, 739 (1947); H. H. Hopkins, Jr., and B. B. Cunningham, Phys. Rev. **73**, 1406 (1948); M. Lindner and I. Perlman, Phys. Rev. **73**, 1124, 1202 (1948); **78**, 499 (1950); H. H. Hopkins, Jr., Phys. Rev. **77**, 717 (1950); L. Marquez, Phys. Rev. **88**, 225 (1952); Miller, Thompson, and Cunningham, Phys. Rev. **74**, 347 (1948); Rudstam, Stevenson, and Folger, Phys. Rev. **87**, 358 (1952); G. Wagner and E. O. Wiig, J. Am. Chem. Soc. **74**, 1101 (1952); S. G. Rudstam, Phil. Mag. **44**, 1131 (1953). See also the reviews by G. T. Seaborg and D. H. Templeton, Ann. Rev. Phys. Chem. **2**, 83 (1951) and G. W. Wilkinson, J. Chem. Soc. **S366** (1949).

[2] R. Serber, Phys. Rev. **72**, 1114 (1947); M. L. Goldberger, **74**, 1269 (1948).

A study has been made of the distribution of radioactive products formed when cesium is irradiated with 240-Mev protons in the Rochester 130-inch cyclotron. Cesium was chosen since it is monoisotopic (Cs^{133}), which enables specific reactions to be established even though the reaction path is not always known.

II. EXPERIMENTAL PROCEDURE

Targets for irradiation consisted of approximately 200 milligrams of spectroscopically-pure cesium chloride[3] which was wrapped in a single sheet of aluminum foil 3.8 cm long, 0.3 cm wide, and 0.2 cm thick, covered by monitor foils 0.001-inch thick. To avoid errors caused by inexactitude in alignment of the target with the beam direction, the monitor and guard foils were placed both before and after the target proper. To minimize the error caused by recoil of Na^{24} out of the monitor foil, a guard foil was placed in front of each monitor so that any loss of Na^{24} by recoil out of the monitor foil would be offset by the gain in Na^{24} from the guard foil. It was found that more accurate measurement of absolute cross sections could be made using a target square in cross section as illustrated in Fig. 1. This minimizes the effect of beam penetration cutoff in the aluminum foil surrounding the target. In this way, it was possible to increase the accuracy of measuring absolute cross sections by a factor of almost 1.5. Measurement of the cyclotron beam current was made using the $Al^{27}(p,3pn)Na^{24}$ reaction in the monitor foils.

After irradiations of about one hour, the target was processed chemically. The aluminum monitor foils were dissolved separately in 1 ml of concentrated hydrochloric acid. After dilution, an aliquot of each of these solutions was mounted on $\frac{7}{8}$-inch filter paper disks backed by 0.005-inch thick stainless steel counting plates one inch in diameter and counted in the standard manner adopted for spallation products under a high-resolution beta-proportional counter. In every case the activities observed were 2.0-hour F^{18}, 15.00-hour Na^{24}, and long-lived activity, presumably a mixture of Be^7 and Na^{22}.

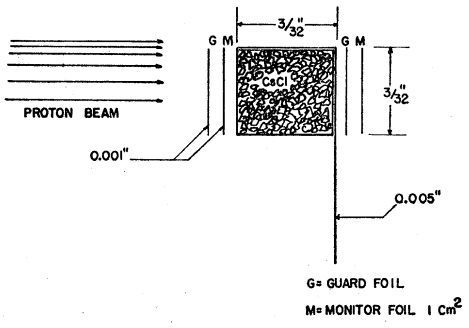

FIG. 1. Cyclotron target, showing placement of 0.001-inch thick guard foils (G) and 0.001-inch thick monitor foils (M) of aluminum. Target wrapping was 0.005-inch aluminum.

The CsCl target itself was dissolved in $6N$ nitric acid for subsequent separation into various elemental fractions. Chemical exchange of radioelements with the inert carriers which were added was assured through vigorous oxidation-reduction steps. The general chemical methods employed were taken, with some modifications,[4] from the fission product procedures discussed by Coryell and Sugarman.[5] In various bombardments the following elements were separated and studied: lanthanum, barium, cesium, xenon, iodine, tellurium, antimony, tin, indium, cadmium, palladium, and ruthenium.

After chemical separation and successive repurifications of the desired elemental fraction, the final precipitate was vacuum filtered in a Hirsch funnel onto a weighed disk of Whatman No. 42 filter paper $\frac{7}{8}$ inch in diameter which had been treated previously with ethanol and dried under a heat lamp. The chemical yield was then determined gravimetrically. The disk containing the active deposit was cemented with Duco to a one-inch diameter stainless steel plate, covered with a thin film of collodion, dried under heat, and counted under a beta-proportional counter, sodium iodide scintillation counter, or beta-ray survey spectrometer.

Yields were computed starting with the raw counting yields obtained from an extremely-stable methane flow beta-proportional counter which is a duplicate of the Los Alamos model.[6] The electronic units were custom-built.[7] The counter consists of an aluminum cathode counting chamber with a $2\frac{1}{4}$-inch diameter window of thickness 4.9 mg/cm² and a 0.002-inch diameter stainless steel central anode wire held at a potential of 4600 volts. Pulses from this chamber pass through 4 feet of RG-13/U coaxial cable to the Model PA-2 pulse amplifier where they are amplified linearly, passed through a trigger circuit after amplification, and then equalized to about 1 volt in amplitude. These are then fed into a fast scaler. The chamber is flushed with pure methane gas[8] and operated at a slow flow rate giving

[3] "Specpure" brand CsCl obtained from Johnson, Matthey, and Company, Ltd., London, Cat. No. J. M. 15. The accompanying spectrographic report lists barely visible lines for Na, Si, and Cu; faintly visible lines of K, Ca, and Mg: and no lines for Ag, Al, As, Au, B, Ba, Be, Bi, Cd, Co, Cr, Fe, Ga, Ge, Hg, In, Li, Mn, Mo, Ni, Pb, Rb, Sb, Sn, Sr, Ti, Tl, V, W, Zn, and Zr.

[4] For detailed chemical separation procedures, see R. W. Fink, Ph.D. thesis, University of Rochester Library, 1953 (unpublished).

[5] C. D. Coryell and N. Sugarman, *Radiochemical Studies: Radiochemistry of the Fission-Product Elements*, (McGraw-Hill Book Company, Inc., New York, 1951 and 1952), National Nuclear Energy Series, Plutonium Project Record, Vol. 9.

[6] E. L. Kemp, Los Alamos Laboratory Report La-1207, 1950 (unpublished); J. H. Larkins, Los Alamos Laboratory Report La-1238, 1951 (unpublished); V. C. Rexroth, Los Alamos Laboratory Report La-1259, 1951 (unpublished).

[7] By Trott Electronics Company, 1944 Clinton Avenue North, Rochester, New York.

[8] "96 percent pure" methane with not more than 4 percent ethane, 2 percent nitrogen, and 0.1 percent oxygen, and sulfur-free. Obtained from Industrial Gas Department, Carbide and Carbon Chemicals Corporation, 30 East 42nd Street, New York 17, New York.

rise to a pressure slightly above atmospheric. With this arrangement, a plateau about 350 volts long with a slope of less than 1 percent per 100 volts was obtained. The resolving time, less than $(0.07\pm0.03)\times10^{-8}$ minute, was determined by using two Y^{91} half-samples which together gave approximately 120 000 counts/minute. By using the decay of a very active source of Na^{24}, it was found that the coincidence correction was negligible up to 500 000 counts per minute. Above this, a small but increasing resolving time correction was found to apply. Saturation set in at 1.3×10^6 counts per minute. In all cases, where the counting rate of the sample was excessive, a lower geometry was used. Because a greater amount of active precipitate can be counted than with a G-M tube, the error in the chemical yield determination can be materially reduced with this instrument since weighing errors become less significant. The counting efficiency for gamma rays, x-rays, and particulate radiation is very similar to that of a standard helium-alcohol G-M tube. All samples were counted on a $\frac{7}{16}$-inch thick shelf of stainless steel and an appropriate estimate of the correction factor for saturation backscattering was applied.

It is worthwhile to give the method and assumptions under which yields were computed from the raw counting yield, A^0, obtained by back extrapolation of the decay curves to the time of cessation of bombardment.

The absolute disintegration rate, N^0, may be related to the observed raw yield, A^0, by the relation

$$N^0=\frac{A^0}{f_C f_W f_A f_B f_{SSA} G E}\ \text{dis/min},$$

where f_C=correction factor for counter resolution (taken as unity for beta-proportional and scintillation counters). f_W=correction factor for total air-window absorption. Under our conditions, the total air-window thickness was about 5 mg/cm². Values of f_W were interpolated from data of Gleason, Taylor, and Tabern[9] for particles of various energies while for electromagnetic radiation a value of 0.99 was used. Where weak conversion electrons are present, this factor may constitute a serious correction. Values for each nuclide were computed as a weighted mean using known or estimated branching ratios for the respective radiations emitted. f_A=correction for air scattering of particles; taken as unity. f_B=correction factor due to backscattering in the source.[10] For saturation backscattering, f_B varies with Z of the backing material. Burtt[11] gives about 1.3 for beta-particles scattered from filter paper or cardboard of infinite thickness. In this work, the following values of f_B were used: for negative electrons, 1.25; for positrons, 1.2; and for electromagnetic radiations, 1.0. That f_B is definitely larger for negatrons than for positrons has been demonstrated by Seliger[12] for all cases. f_H=correction factor for scattering of particles from the housing (taken as unity for the Lucite and aluminum housing used). f_{SSA}=correction factor for effect of self-scattering and self-absorption owing to the finite source thickness. For each nuclide in each individual sample, a value of this factor was interpolated from the data of Nervik and Stevenson.[13] For electromagnetic radiations, f_{SSA} was assumed to be unity. In cases of nuclides having known branching ratios between particles of various energies and electromagnetic radiations, or where these ratios could be estimated from absorption curves in aluminum, a weighted mean value was calculated. This factor becomes the more serious the weaker the particle energy and the thicker the sample. G=correction factor for geometry. G is the solid angle fraction subtended by the sensitive volume of the counter. The value of G was calculated from geometrical measurements as 0.347, while from an absolutely-calibrated Co^{60} source, assuming 2 percent efficiency for Co^{60} gamma rays, the value was 0.316. For all corrections used here, G was taken as 0.32. E=correction factor for counting efficiency, defined as the ratio of the number of events recorded by the counter to the total number of particles or photons traversing the sensitive volume of the counter. This factor constitutes the largest over-all error to the determination of absolute cross sections. In order to establish the true counting efficiency for x-rays, samples of Fe^{55} and Cs^{131} were prepared and taken to Brookhaven National Laboratory[14] where they were calibrated using a high-pressure argon-methane x-ray proportional chamber and pulse analysis equipment. These calibrated x-ray standards were subsequently used to measure the x-ray counting efficiency of the beta-proportional counter and a helium-alcohol G-M Tube. The experimental results are shown in Table I. In this work, E was taken as 0.25 percent for 30 kev x-rays, 1 to 2 percent for high-energy gamma

TABLE I. Experimental values for counting efficiencies of xenon and manganese x-rays.

Detector	Values in percent: Xenon K x-rays (30 kev)	Manganese K x-rays (6.4 kev)
Methane beta-prop. counter	0.25	0.8_3
Tracerlab TCG-2 GM tube, He-alcohol	0.1_5	0.2

[9] Gleason, Taylor, and Tabern, Nucleonics **8**, No. 5, 12 (1951).
[10] See L. Yaffe and K. M. Justus, J. Chem. Soc. **S341** (1941).
[11] B. P. Burtt, Nucleonics **5**, No. 2, 28 (1949).
[12] H. H. Seliger, Phys. Rev. **78**, 491 (1950); Phys. Rev. **88**, 408 (1952).
[13] W. E. Nervik and P. C. Stevenson, Nucleonics **10**, No. 3, 18 (1952).
[14] We appreciate the hospitality extended to us by the Brookhaven National Laboratory and assistance with the equipment provided by Dr. M. L. Perlman, of the Department of Chemistry.

TABLE II. Experimental results of 240-Mev Cs-p-spallation studies.

Nominal half-life	Nuclide	Assumed counting efficiency, E	Average absolute cross section in millibarns
15.0 hr	Na^{24}	1.0	11.0 (Monitor)
2.4 days	Ba^{128}	1.0	8.1
2.0 hr	Ba^{129}	1.0	3.6
12.0 days	Ba^{131}	0.72	5.3
5.5 hr	Cs^{127}	1.0	4.5
31 hr	Cs^{129}	0.39	15
9.8 days	Cs^{131}	0.0025	460
7.1 days	Cs^{132}	0.005	117
1.6 hr	$I^{120+121}$	1.0	11
13 hr	I^{123}	0.17	21
4.0 days	I^{124}	0.30	8.4
12.5 days	I^{126}	0.53	5.0
2.5 hr	Te^{117}	1.0	2.4
6.0 days	Te^{118}	1.0	4.8
16 hr	Te^{119}?	0.002?	50?
17.0 days	Te^{121}	0.09	?
90 days	Te^{121m}?	1.0	0.3
2.8 hr	Sb^{117}	0.15?	4.2
6.0 days	Sb^{120}	0.1?	4
2.6 days	Sb^{122}	1.0	0.3
60 days	Sb^{124}	1.0	0.07
4.5 hr	Sn^{108}	0.05?	0.3
2.0 hr	$In^{108,110}$	1.0	0.02
4.2 hr	In^{109}	0.9	0.05
6.7 hr	Cd^{107}	0.05	0.9
50 min	$Cd^{104,105}$	1.0	0.2
4.0 days	Pd^{100}	0.05	1.5
1.7 hr	Ru^{95}	1.0	0.2
2.8 days	Ru^{97}	0.07?	3?
4 hr	La^{133}?	1.0	0.005?

rays, and 99 percent for particulate radiation. The actual counting efficiency assumed for each nuclide is listed with the final results in Table II.

To convert the absolute counting rate N^0 into an absolute cross section, it is necessary to correct each yield to saturation bombardment. By assuming a constant rate of production in the cyclotron, we obtain the relation

$$N_\infty^0(A)=N^0(A)/(1-e^{-\lambda_A T}),$$

where $N_\infty^0(A)$ is the yield of species A at infinite bombardment, T is the actual duration of irradiation, and λ_A is the decay constant of species A. Since I^{126} was the only species determined in all bombardments, it was chosen as the internal monitor to which all other yields were compared. To convert these relative yields into absolute cross sections, it is first necessary to evaluate the absolute cross section of the internal monitor I^{126} by comparison of its yield with that of Na^{24} from the $Al^{27}(p,3pn)Na^{24}$ reaction[15] assuming thin target formulation. The result is

$$\frac{\sigma(I^{126})}{\sigma(Na^{24})}=\frac{N_\infty^0(I^{126})}{N_\infty^0(Na^{24})}\times f,$$

where $f=6.24\times$(weight of aluminum monitor/weight of CsCl target). The factor f corrects for the fact that the total number of cesium atoms exposed to the proton beam is different from the total number of atoms of aluminum monitor. By this method, the absolute cross section for formation of I^{126} from cesium at 240 Mev is 5.0 millibarns.

III. DISCUSSION OF ERRORS

In addition to the error in the absolute cross sections due to errors in interpolating values of absolute counting correction factors, chemical yields, counting efficiencies, resolution of decay curves having many components, which lead to an estimated total indeterminate error of from 20 percent to 60 percent in general, there are some intangible effects which cannot be evaluated directly.

These effects are associated with the fact that in most cases one is dealing with short lived, highly neutron-deficient nuclides whose yield may be partly primary and partly the result of the decay of very short-lived parent nuclides produced as primary spallation products. For example, the cross sections of $I^{120+121}$, Te^{117}, Cs^{127}, and other neutron-deficient nuclides having unknown or very short-lived parent nuclides, may quite possibly be excessively large due to contributions from short-lived parents. It is clear that yields of those species subject to this effect are much more susceptible to such bombardment conditions as intermittent beam intensity fluctuations and variations in the duration of bombardment. Indeed, from bombardment to bombardment, the yields of I^{124}, I^{126}, Cs^{132}, and other shielded nuclides appear to be smoother and more reproducible than of such species as $I^{120+121}$, Te^{117}, Cs^{127}, and other similar species. It is known,[16] for example, that Ba^{127} has a 12-minute half-life. Thus, during the 2 to 3 hours required for chemical separation, this nuclide decays out before the barium fraction can be separated from the cesium fraction. What the independent yield of Cs^{127} is, therefore, has not been determined.

Errors can be caused by lack of radiochemical purity. In this work, the purity of the various elemental fractions was satisfactory as evidenced by decay curves and beta-ray spectrometer spectra. In the case of the

[15] Values of the cross section for $Al^{27}(p,3pn)Na^{24}$ were interpolated at 240 Mev from data of V. Peterson *et al*, University of California Radiation Laboratory file compiled by W. W. Meinke, Jr., 1950 (unpublished). Values are also published by N. H. Hintz and N. F. Ramsey, Phys. Rev. **88**, 19 (1952), and by L. Marquez, Phys. Rev. **88**, 225 (1952).

[16] M. Lindner and R. N. Osborne, Phys. Rev. **88**, 1422 (1952).

lanthanum fraction, however, that yield assigned to 4-hour La^{133} may be due entirely to the carrying of 4-hour In^{109} or Sn^{108} even through the many steps and repurification procedures used. Although the question of whether La^{133} was formed by a secondary $(\alpha,4n)$ reaction, not previously observed in spallation, is interesting, it has not been established in this study.

IV. INTERPRETATION OF RESULTS

The experimental values of the absolute cross sections of various nuclides formed when cesium is bombarded with 240-Mev protons[17] are given in Table II.

These data indicate that the yield pattern of radioactive spallation products is as follows.

1. From mass numbers 131 to 127, a loss of up to 6 nucleons, the absolute cross sections drop by a factor of 50. The largest yields by far are represented by mass numbers 131 and 132, one and two neutrons removed from the target. Presumably this reflects the primary knock-on reaction whereby the incident proton interacts with only one or two nucleons according to the mechanism of Serber and Goldberger.[2] The struck nucleons are then emitted in the order of 10^{-22} second as high-energy secondaries sharply peaked in the forward direction. Forward peaked neutrons and charged particles[18] have been observed in high-energy spallation reactions. At an energy as high as 240 Mev, the contribution of the neutron "pickup" reactions (p,d) and (p,t) would not be expected to account appreciably for the very large yields of Cs^{132} and Cs^{131}; rather, it would be expected that the instantaneous knock-on process caused by glancing collisions in which only a small excitation energy is imparted to the struck nucleus would be the responsible mechanism.

2. From mass numbers 127 to 120, a further loss of up to 7 nucleons from the excited nucleus, the yields are fairly constant and of the order of 10 millibarns. This constancy of yields reflects the fact that the residual excitation energies left to the struck nuclei after the primary knock-on process are broader in distribution and generally much larger than in typical low-energy cases where the Bohr-Wheeler compound nucleus mechanism[19] prevails. The wider and higher distribution of excitation energies results in a broad distribution of end products, a phenomenon which is in contrast to the narrow distributions prevailing in low-energy reactions.

3. From mass numbers 120 to 110, a further loss of up to 10 nucleons, the yields decrease by another factor of 10 and continue to decrease at the same rate down to mass number 95, the lowest measured. At still lower mass numbers, the yield curve should rise again because of the contribution from the competing process of fission. The study of cesium fission has not been included in the scope of this work.

4. Ordinarily, in a chemical study it is not possible to ascertain whether the nucleons ejected come off singly or in fragments. The fact, however, that such products as Ru^{95}, Pd^{100}, and Cd^{107} are found chemically leads to the conclusion that fragmentation must play an important role in the evaporation process. For the formation of Ru^{95}, Pd^{100}, or Cd^{107} from cesium by evaporation of single nucleons, the energetic requirement is much larger than the actual bombardment energy of 240 Mev. Thus, only by the emission of alpha particles and larger fragments can one account for the presence of Ru^{95}, Pd^{100}, Cd^{107}, etc., among the end products. Evidence for alpha-particle emission has been given by Wagner and Wiig,[1] while heavier fragments have been identified by Wright[20] and by Marquez and Perlman.[21]

Yields of those elements more likely to be formed through the evaporation process following the initial knock-on step are accentuated toward the most neutron-deficient nuclides. This is true for iodine, tellurium, antimony, and possibly barium nuclides, the latter arising from neutron evaporation from excited Ba^{133} formed by the (p,n) charge exchange reaction. It is decidedly not true for cesium isotopes whose yield pattern is mostly due to the primary knock-on process. Accentuation of the yields of the most neutron-deficient species is more marked the higher the energy and the heavier the target element as expected from the increase in the Coulomb barrier with increasing atomic number.

These data are in agreement with the general pattern for all cases studied. About 80 percent of all interactions leads to nuclides in the vicinity of the target element, followed by respectable and comparatively constant yields to about 15 or 16 mass numbers below the target, and finally a sharp and continuing drop in yields until the fission product region is reached where the yields are expected to rise again.

V. ACKNOWLEDGMENTS

We are happy to acknowledge the cooperation of Professor S. W. Barnes, Mr. William Coombs, Mr. Kurt Enslein, and the crew of the Rochester 130-inch cyclotron.

[17] Results of excitation studies of 60 to 240-Mev protons on cesium will appear in a forthcoming paper.

[18] J. Hadley and H. York, Phys. Rev. **75**, 1467 (1949); Phys. Rev. **80**, 345 (1950).

[19] N. Bohr, Nature **137**, 344 (1936); Science **86**, 161 (1937); N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[20] S. C. Wright, Phys. Rev. **79**, 838 (1950).

[21] L. Marquez and I. Perlman, Phys. Rev. **81**, 953 (1951).