

## The Transmutation of Fluorine by Protons

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The excitation functions for the production of long range alpha-particles, gamma-rays, and electron pairs by the bombardment of  $F^{19}$  by protons have been observed simultaneously up to a bombarding energy of 1.5 Mev. The long range alpha and pair curves exhibit resonance peaks superimposed on a background of increasing intensity with increasing bombarding energy. Approximate coincidence in two instances of pair and alpha-resonances suggests that full range alphas and the short range alphas preceding pair emission can be products of competing

modes of decay of the same intermediate states of  $Ne^{20}$ . This in turn suggests that the state of  $O^{16}$  which decays by pair emission has the same parity (even) as the ground states of  $O^{16}$  and thus that the pair emission can be due to ordinary electromagnetic forces. The absolute yields of the various processes have been measured and a discussion of the measurement of high energy gamma-ray and pair yields by means of electroscopes is given. The large ratio of gamma-ray yields to long range alpha and pair yields previously observed has been confirmed.

### INTRODUCTION

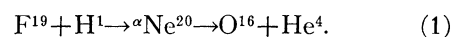
THE transmutation of fluorine by protons has been of considerable experimental and theoretical interest from the very beginnings of nuclear research employing particles accelerated to high velocities by modern electrical methods. The abundant yield of the reactions resulting from the bombardment of fluorine by protons and the fact that naturally occurring fluorine consists of only one isotope (at least to within 1 percent) have been of great advantage to the experimental nuclear physicist. Excited states of  $Ne^{20}$  are the intermediate products of the bombardment, while states of  $O^{16}$  along with the ground state of  $He^4$  are the final products in the most important cases. Knowledge of the states of these "completed shell" or "alpha-model" nuclei with their particular properties of symmetry in neutrons and protons will be of great importance in the development of an adequate theory of nuclear structure.

### The long range alpha-particles

In their report of their early work on disintegration by artificially accelerated particles Cockcroft and Walton<sup>1</sup> announced the production of alpha-particles in the bombardment of fluorine with protons. Similar observations were made by Oliphant and Rutherford.<sup>2</sup> The range of the alpha-particles was less than that observed by

later investigators and it is possible that they were spurious, being due to some contaminant.<sup>3</sup> The first observation of alpha-particles which has been consistently verified was by Henderson, Livingston and Lawrence.<sup>3</sup> These observers reported a single alpha-particle group of range 6.7 cm; this was obtained with protons of 1.2 Mev energy. Burcham and Smith<sup>4</sup> have since made a more careful determination of the range; with a proton energy of 0.85 Mev, the alpha-particle range in a direction perpendicular to the beam was 5.90 cm, which corresponds to an energy release of 7.95 Mev; again only a single group was found.

It is now certain that the origin of these alpha-particles is the following reaction:



In this equation, the superscript  $\alpha$  designates a particular kind of excited state in the intermediate  $Ne^{20}$  nucleus which may decay with long range alpha-emission corresponding to the full energy available in the reaction.

In their experiment, Henderson *et al.* found a rapid increase in the yield as the proton energy was increased from 0.7 to 1.5 Mev. Burcham and Devons<sup>5</sup> made a more precise investigation of the excitation curve for proton energies from 0.53 to 0.93 Mev; in addition to a continuous increase

<sup>1</sup> J. D. Cockcroft and E. T. S. Walton, Proc. Roy. Soc. **A137**, 229 (1932).

<sup>2</sup> M. L. E. Oliphant and Lord Rutherford, Proc. Roy. Soc. **A141**, 259 (1933).

<sup>3</sup> Henderson, Livingston and Lawrence, Phys. Rev. **46**, 38 (1934).

<sup>4</sup> W. E. Burcham and C. L. Smith, Proc. Roy. Soc. **A166**, 176 (1938).

<sup>5</sup> W. E. Burcham and S. Devons, Proc. Roy. Soc. **A173**, 555 (1939).

in yield with increasing bombarding energy, they found two resonances at 0.72 and 0.83 Mev.

#### Gamma-radiation and the short range alpha-particles

McMillan<sup>6</sup> first observed gamma-radiation produced in the bombardment of fluorine with protons. The earliest estimates of the energy of this radiation were based on measurements of its absorption in lead. They were made by McMillan<sup>7</sup> and by Crane, Delsasso, Fowler and Lauritsen,<sup>8</sup> who employed ionization chambers and electroscopes to measure the intensity. The value of the absorption coefficient in lead,  $0.49 \text{ cm}^{-1}$  gave, according to the Klein-Nishina theory of the Compton effect, a quantum energy of 2 Mev. However, the origin of such a gamma-ray could not be accounted for. Oppenheimer suggested that the inferred value of the energy was too low, and that the absorption was due not only to the Compton effect, but also to pair production. Measurements of the absorption in other elements, tin, copper and aluminum, showed that this suggestion was correct; the results obtained were all consistent with, and uniquely determined the value of approximately 5.4 Mev for the quantum energy.

The technique of these measurements is open to serious criticism.<sup>9</sup> The radiation removed from the beam by the absorber is partly replaced by a secondary radiation whose presence makes the interpretation of the measurements difficult. This objection is eliminated in a method introduced and employed by Delsasso, Fowler and Lauritsen.<sup>10</sup> The gamma-radiation ejects pairs from a thin lead radiator in a cloud chamber; on alternate expansions the beam is passed through the absorbing body; the energy of each pair can be determined from the curvature in a magnetic field of the paths of the two members with sufficient accuracy to decide whether or not it was produced by the primary gamma-ray. The transmission of the absorber is the ratio of the number of full energy pairs produced with the ab-

sorber in the beam to the number produced with the absorber removed. In this way the value  $0.4 \pm 0.1 \text{ cm}^{-1}$  was obtained for the absorption coefficient in lead. Using this same method Halpern and Crane<sup>11</sup> found the absorption coefficient in aluminum to be  $0.062 \pm 0.009 \text{ cm}^{-1}$ . These values are consistent with theoretical calculations of the absorption coefficients made from direct determinations of the energy of the gamma-ray.

The energy was determined directly by Crane, Delsasso, Fowler and Lauritsen<sup>8</sup> from the spectrum of positive and negative electrons ejected by the gamma-radiation from a thick lead sheet in a cloud chamber; the electron energies were deduced from the path curvature due to a known magnetic field. The value obtained for the quantum energy was 5.4 Mev.

This method was later modified,<sup>10</sup> and a thin lead radiator was used. This made it possible to associate the two members of a pair and to determine their energies without having the uncertainty of an appreciable energy loss in the material. The value obtained for the quantum energy was  $6.0 \pm 0.2$  Mev. As described in the preceding paper of this issue of *The Physical Review*, Lauritsen, Lauritsen and Fowler have found, by a more careful application of this method, the value  $6.2 \pm 0.1$  Mev.

By measuring with coincidence counters the absorption in aluminum of the secondary electrons ejected by the gamma-ray, Curran, Dee and Petržílka<sup>12</sup> arrived at values of 6.3 and 5.5 Mev for the quantum energy, according as they based their conclusions on the maximum range in the absorber or the half-value thickness. The source of the discrepancy between the two values is difficult to understand as the radiation has not been found to be inhomogeneous as supposed by Curran *et al.* It must be pointed out however that the secondary absorption method must be calibrated by direct determinations of the quantum energy. The work of Dee, Curran and Strothers<sup>13</sup> with a magnetic spectrograph gave 6.5 Mev.

<sup>6</sup> E. McMillan, Phys. Rev. **46**, 325 (1934).

<sup>7</sup> E. McMillan, Phys. Rev. **46**, 868 (1934).

<sup>8</sup> Crane, Delsasso, Fowler and Lauritsen, Phys. Rev. **46**, 531 (1934).

<sup>9</sup> Delsasso, Fowler and Lauritsen, Phys. Rev. **51**, 391 (1937).

<sup>10</sup> Delsasso, Fowler and Lauritsen, Phys. Rev. **51**, 527 (1937).

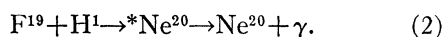
<sup>11</sup> J. Halpern and H. R. Crane, Phys. Rev. **55**, 258, 260 (1939); E. R. Gaertner and H. R. Crane, *ibid.* **52**, 582 (1937).

<sup>12</sup> Curran, Dee and Petržílka, Proc. Roy. Soc. **A169**, 269 (1938).

<sup>13</sup> Dee, Curran and Strothers, Nature **143**, 759 (1939).

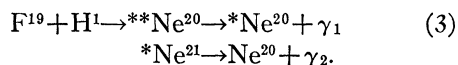
Long before the origin of the gamma-radiation was understood, its pronounced resonance character had been demonstrated by various investigators.<sup>12, 14-16</sup> The most extensive work was by Bernet, Herb, and Parkinson.<sup>17</sup> Their results showed that most of the resonance levels are very narrow: in fact, the observed widths of most of the peaks were thought to be experimental. The low energy part of the curve was investigated more closely by Burcham and Devons.<sup>5</sup> They were able to reduce the observed widths of the peaks at 0.33 and 0.67 Mev to 6 kev, and presumably this is still chiefly experimental. However, the level at 0.59 Mev was found to have a width of 35 kev.

The simplest reaction which has been proposed for the origin of the gamma-radiation is:



From the known masses the quantum energy is expected to be  $13.1 \pm 0.3$  Mev for an 0.33-Mev proton. The discrepancy between this and the observed value definitely rules out this simple process.

The fact that the calculated value is roughly twice the observed energy of the radiation suggested a cascade process in which two quanta of approximately equal energies are emitted:

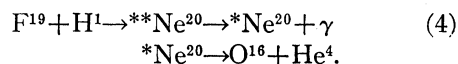


To detect the simultaneous emission of two quanta which this proposal implies, Dee, Curran and Strothers<sup>13</sup> connected two gamma-ray counters in a coincidence circuit; coincidences occurred with a frequency less than 1 percent of that to which the proposed process would lead.

This hypothesis was subjected to additional tests. According to reaction (3) a change in proton energy should be accompanied by a modification of the gamma-ray spectrum; presumably the energy of the first quantum,  $\gamma_1$ , should increase by 19/20 of the increase in proton energy. However, when the bombarding voltage

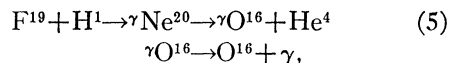
was raised from 0.330 to 0.86 Mev, the gamma-ray spectrum, as indicated by the maximum range of its secondary electrons in aluminum,<sup>13</sup> measured with coincidence counters, was constant to within 0.05 Mev. The same conclusion was reached when the gamma-ray secondaries were measured with a magnetic spectrograph; no change greater than  $\pm 0.1$  Mev was observed. A similar result was found by Lauritsen, Lauritsen and Fowler whose results are given in Table I of the preceding paper.

Another possibility which has been considered is the following:



But the objections given in the preceding paragraph will also apply here and this proposal is not tenable.

The now accepted explanation of the origin of the gamma-rays is the following:



where the superscript  $\gamma$  refers to states of  $Ne^{20}$  or  $O^{16}$  involved in the production of the 6.2-Mev gamma-radiation. According to the masses and the gamma-ray energy these alpha-particles should have somewhat less than 1 cm range. No such group of alpha-particles had been observed in the earlier work on alpha-particle production. Owing to the presence of scattered protons this would have been impossible except at low bombarding energies. An unsuccessful search for this short range group had been made by Burcham and Smith.<sup>4</sup> However, at the time of their work the energy of the gamma-radiation was thought to be 5.7 Mev, and using this figure they calculated for the range of the alpha-particles a value which is now known to be too large. Under the conditions of their experiment—0.85 Mev bombarding voltage—the range of the alpha-particles is still less than that of the scattered protons.

When the higher values for the gamma-ray energy were obtained, the search for the short range alpha-particles was renewed. Calculations based on the new data indicated that only at proton energies less than 0.5 Mev would the range of the alpha-particles exceed that of the

<sup>14</sup> Hafstad, Heydenburg and Tuve, Phys. Rev. **49**, 866 (1936).

<sup>15</sup> Hafstad, Heydenburg, and Tuve, Phys. Rev. **50**, 504 (1936).

<sup>16</sup> Herb, Kerst and McKibben, Phys. Rev. **51**, 691 (1937).

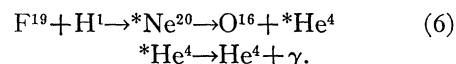
<sup>17</sup> Bernet, Herb and Parkinson, Phys. Rev. **54**, 398 (1938).

protons. These short range alpha-particles were first observed by McLean, Becker, Fowler and Lauritsen<sup>18</sup> who used a proton energy of 0.35 Mev. These observers also showed that this production of alpha-particles displayed a resonance between 0.30 and 0.35 Mev. Such a resonance was already known to exist in the production of gamma-radiation, and the association of the alpha- and gamma-rays seems quite certain. Similar results were obtained almost simultaneously by Burcham and Smith.<sup>19</sup> Burcham and Devons<sup>5</sup> extended this work by showing that the excitation function for the short range alpha-particles is identical with that of the gamma-radiation in the proton energy range from 0.30 to 0.95 Mev. This excitation function has a very striking structure which makes the comparison quite definite. In this work it was necessary to deflect the scattered proton beam by means of a strong magnetic field. Their resolution, particularly in the alpha-particle measurements, was not good, but there is an obvious correspondence between the prominences of their curve and the six known gamma-ray resonances which lie in this energy range. From the range of the alpha-particles,  $0.86 \pm 0.05$  cm, and the masses involved, McLean *et al.* calculate the  $Q$  of reaction (5) to be  $1.74 \pm 0.10$  Mev. This leads to the value  $6.2 \pm 0.2$  Mev for the gamma-ray energy. This is in agreement with the best direct determinations. This indirect measurement should give the most reliable value. Within the experimental error, the same value was obtained by Burcham and Devons. These investigators also showed that at the 0.33, 0.66, and 0.87 Mev resonances the differences of the alpha-particle energies is  $\frac{3}{4}$  of the corresponding differences in bombarding energy.\* This is the result predicted on the basis of reaction (5). On the other hand, if we assume, as implied in reaction (4), that the alpha-particles always originate in the same transition, whatever the bombarding voltage, we would expect the energy of those alpha-particles

emitted perpendicularly to the beam to decrease by 1 percent of the increase of proton energy.

These results, together with the previously mentioned observations on the gamma-ray spectrum, provide very conclusive evidence that reaction (5) describes the production of the gamma-radiation at the 0.334, 0.479, 0.589, 0.660, 0.862, 0.927, 1.335 and 1.363 Mev resonances. These resonances contribute approximately 75 percent of the gamma-radiation produced below 1.5 Mev bombarding energy.

Of course, there also is the possibility of a reaction similar to reaction (5) in which the alpha-particle is formed in an excited state, and subsequently emits a photon:



However, no state of the helium nucleus so near to the ground state as is here required by the quantum energy has been suggested by the many reactions in which alpha-particles are produced.

#### The production of electron pairs

In the investigations described up to this point no soft gamma-radiation had been discovered; in fact, in all of the experiments, a small amount of filtering was employed to exclude the soft characteristic x-radiation emitted by all targets under proton bombardment.<sup>20</sup> Using a thin wall target tube and a thin wall electroscopes, Fowler and Lauritsen<sup>21</sup> found a soft radiation in addition to the characteristic x-radiation. To establish its energy, absorption measurements in lead and aluminum were made at 0.82 and 1.13 Mev bombarding energy. These indicated that the radiation consists of electrons, not gamma-rays as first supposed. This conclusion was checked by cloud-chamber observations made with 0.82 Mev bombarding energy which showed the radiation to consist of electron pairs with total energy estimated as  $5.9 \pm 0.5$  Mev.

By using two electroscopes, one shielded with  $\frac{1}{8}$  inch of lead to detect only gamma-radiation, and the second unshielded except for target tube

<sup>18</sup> McLean, Becker, Fowler and Lauritsen, Phys. Rev. **55**, 797 (1939).

<sup>19</sup> W. E. Burcham and C. L. Smith, Nature **143**, 796 (1939).

\* The incorrect value  $\frac{4}{3}$  for this ratio was published. The experimental values agree more closely with the correct value  $\frac{3}{4}$ .

<sup>20</sup> Livingston, Genevese, and Konopinski, Phys. Rev. **51**, 835 (1940).

<sup>21</sup> W. A. Fowler and C. C. Lauritsen, Phys. Rev. **56**, 840 (1939).

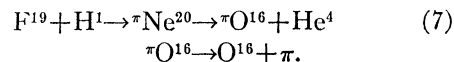
and electroscopes walls to record both gamma-rays and pairs, it was possible to observe simultaneously the excitation functions of these two types of radiation. Their results show that the pair formation as well as the gamma-radiation displays sharp resonances, but the two sets of resonances do not coincide. These are apparently the same pairs observed by Gaertner and Crane<sup>11</sup> who, working with an alternating voltage supply, had made no attempt to determine the excitation functions of the pairs and the gamma-rays.

The difference between the excitation functions of these two kinds of radiation indicates that the pairs do not originate in the process of ordinary pair internal conversion. For example at 1.22-Mev proton energy, there is a peak in the pair excitation but not in the gamma-excitation and at this point the intensity of the pairs is 30 percent of the gamma-ray intensity. Theoretically the pair internal conversion coefficient for any multipole order radiation is less than one-half of one percent.<sup>22</sup> An apparently satisfactory explanation of this unusual process is that the lowest excited state of  $O^{16}$  has angular momentum  $J=0$ ; then decay to the ground state  $O^{16}$  which is known to have angular momentum  $J=0$  cannot take place with the emission of a single quantum. Calculations of Oppenheimer and Schwinger<sup>22</sup> show that pair formation under these conditions is more probable than the emission of two quanta. The questions involved in this calculation are mentioned below.

One must also consider the possibility that the pairs are formed by transition in the  $Ne^{20}$  nucleus. But the existence of many low lying levels in this nucleus<sup>23</sup> makes it unreasonable to expect that gamma-transitions from any state 5.9 Mev above the ground state are rigorously forbidden.

Recently in this laboratory Becker, Fowler, and Lauritsen (unpublished) have observed a group of short range alpha-particles associated with the prominent pair resonance at 1.22 Mev. The alpha-particles were magnetically separated from the scattered protons. The pairs may thus

with some certainty be attributed to the reactions:



In accounting for the pair formation it was necessary to assume the state  $\pi O^{16}$  had angular momentum zero, but the parity was not specified. It would be interesting to know the parity of this state. For if the parity is odd, pair formation can take place only as a result of a non-electromagnetic coupling between the nuclear particles and the pair field such as is postulated in the Gamow-Teller theory of nuclear forces. On the other hand, if the parity is even, pair emission can occur as a result of ordinary electromagnetic forces. If it could be shown that transitions to the states  $\pi O^{16}$  and  $O^{16}$  from the same level in  $Ne^{20}$  occur, this would establish the parity of  $\pi O^{16}$  as even, since normal  $O^{16}$  certainly has even parity: a negative result would be an argument for odd parity.<sup>22</sup> The experiment to be described was undertaken to see if such information could be obtained from the excitation curves in the region of higher voltages. Estimates of the yields of the various reactions were also made.

#### EXPERIMENTAL PROCEDURES

The source of high velocity protons was the pressure electrostatic generator and accelerating tube built and used by Lauritsen, Lauritsen and Fowler and described in the preceding paper. The tube voltage was measured with a generating voltmeter and was checked during every run at some pronounced resonance in the gamma-ray excitation curve. The experimental fluctuation in the energy of the ion beam was not as small as has been secured by Herb *et al.*,<sup>16, 17</sup> but was small enough to permit good resolution of the known gamma-ray resonances.

Some unsteadiness in the ion current made necessary an integrating device to measure the total charge carried by the bombarding protons during a run. The target, at the bottom of a Faraday cage, was bombarded by the beam defined by an aperture slightly above the entrance of the cage, and the charge accumulated by the cage was let into a condenser having good insulation, whose potential could be read continuously on an electroscopes connected across it.

<sup>22</sup> J. R. Oppenheimer and J. S. Schwinger, Phys. Rev. 56, 1066 (1939).

<sup>23</sup> T. W. Bonner, Proc. Roy. Soc. A174, 339 (1940).

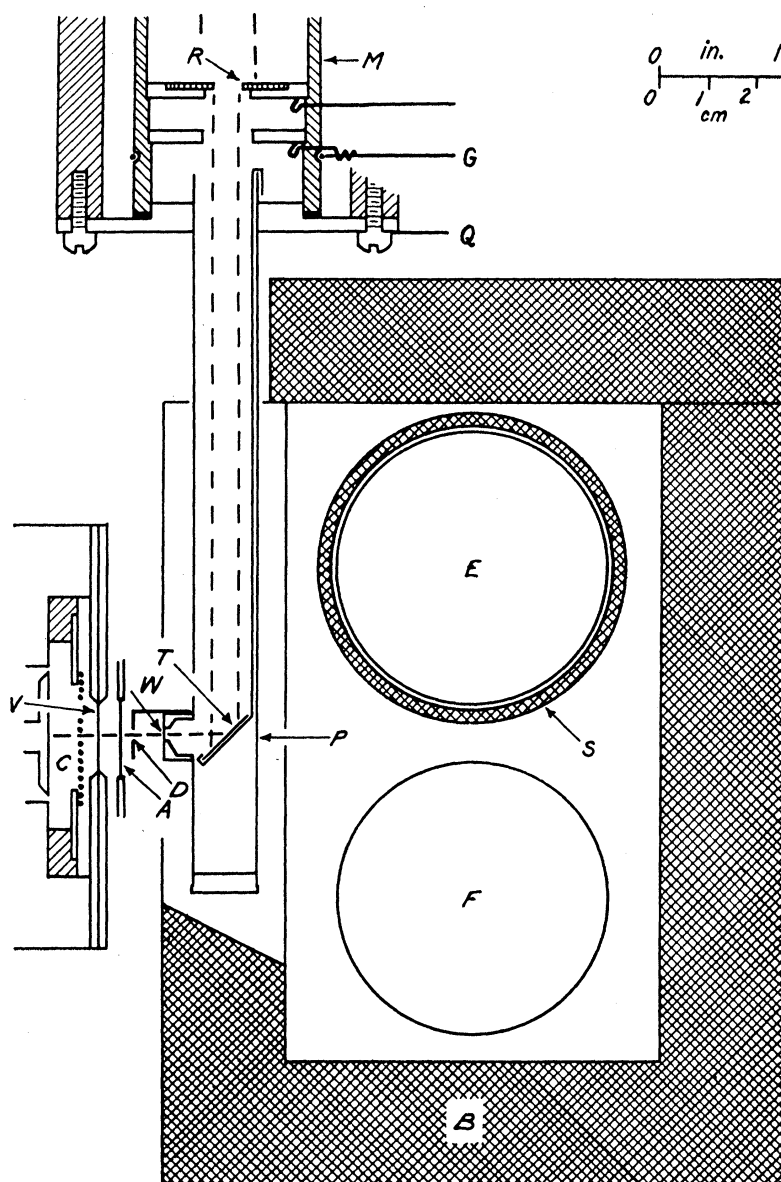


FIG. 1. Arrangement of target tube, electroscopes and ionization chamber. The vertical proton beam, shown by dotted lines, is defined by the 0.25-inch hole in the quartz ring *R*. In aligning the target tube, the position of the beam is observed through the transparent Plexiglas tubing *M* by the fluorescence produced in the quartz ring. The connection *Q* is to the current integrator. A negative potential on *G* prevents erroneous current measurements resulting from a gain or loss of secondary electrons. The lead box *B* completely encloses the electroscopes *E* and *F* except for holes admitting the microscope tubes, holes for illumination, and the beveled slot just large enough to accommodate the target tube *P*. *S* is the 0.125-inch lead shield which absorbs electrons originating in the target. Electrode *E* records only gamma-radiation. Electrode *F* records both electrons and gamma-radiation. The material between the target and this electrode is: the target backing, *T*=0.0010 inch of tantalum, the target holder=0.008 inch of phosphor bronze, the target tube=0.004 inch of German silver, and the electroscopes wall=0.030 inch of aluminum. The path of an alpha-particle emitted perpendicularly to the proton beam is shown by a dotted line. The particle leaves the tube through the 0.00025-inch aluminum window *W*, and enters the ionization chamber *C* through the similar window *V*. The alpha-particle beam is limited by the aperture *D*. Because of the large window *V*, the position of the ionization chamber is not critical. The aluminum absorber *A* stops scattered protons.

This apparatus was calibrated by observing the electroscopes deflection produced by a measured current flowing into the condenser for a measured time. Measurements of the ion current were always made with the condenser negatively charged so that the secondary electrons formed at the defining aperture were repelled from the cage.

It was found that a thin target of  $\text{TaF}_5$  formed by the action of hydrofluoric acid on tantalum as described by Bernet *et al.*,<sup>17</sup> and then

polished, gave more dependable results than the targets formerly used in this laboratory made by depositing a thin layer of  $\text{CaF}_2$  on a copper backing. Two thin target curves were made. Before the second set of data was taken, the target was well polished; being thinner, it then gave a smaller yield, but permitted greater resolution. The thick target was a heavy deposit of  $\text{CaF}_2$  on a copper backing.

A formula has been derived<sup>17</sup> for estimating the thickness of thin targets from a comparison

of the thin target and thick target yield curves. The thickness in energy units is given by the ratio of the area under the thin target resonance peak to the corresponding step in the thick target yield curve. For measurements of this kind it is convenient to use the two resonances at 0.862 and 0.927 Mev for these close peaks are very much more intense than the background upon which they are superimposed. In this way the thickness of the thin target was found to be 8 kev for 1-Mev protons during the first observations and 1.4 kev after polishing.

#### The measurement of the radiations

The possibility of uncertainties in the voltage made it necessary to observe all of the radiations simultaneously. Otherwise resonances in two different radiations occurring close together could not definitely establish the radiations as competing modes of decay of a single level of the compound nucleus. The arrangement of the measuring apparatus is shown in Fig. 1.

The gamma-rays were recorded in a Lauritsen electroscope shielded by  $\frac{1}{8}$  inch of lead. The combined effect of the pairs and gamma-rays was recorded in a similar unshielded electroscope. This is the arrangement previously employed by Fowler and Lauritsen.<sup>21</sup> The two electroscopes were placed as close as possible to the target. Within convenience the material between the target and the unshielded electroscope was reduced to a minimum to avoid absorption of the pairs. To decrease the background due to undesired radiation, principally x-rays and gamma-rays originating from ion bombardment of parts of the tube, the electroscopes were almost completely enclosed in a lead box with one-inch walls.

The target was inclined  $45^\circ$  with respect to the beam, and long range alpha-particles ejected at right angles to the beam passed through a thin aluminum window, whose stopping power, measured with the alpha-particles from polonium, was equivalent to 1.8 cm of air. They passed through 1 cm of air, entered the ionization chamber through a similar window, and were counted by means of a four-stage linear amplifier and thyratron recording circuit; the performance of this apparatus was frequently checked with a polonium alpha-particle source. A diaphragm to

limit the alpha-particle beam was located between the two windows. This usually was a  $\frac{1}{8}$ -inch circular hole  $\frac{3}{4}$  inch from the center of the target, but was sometimes changed to accommodate the large variations in intensity over the voltage range investigated. The ionization chamber was placed in a position convenient for counting the alpha-particles produced by the low energy protons; then when the voltage was raised above 1.1 Mv scattered protons were able to enter the chamber. It would have been possible to move the ionization chamber farther from the tube so as to count only the alpha-particles, whose range is also increased, but to avoid disturbing the geometrical arrangement it was thought better to insert in front of the chamber aluminum foils to absorb the protons when using potentials over 1 Mv; the number of absorbers was varied with the voltage. The correct stopping power of the absorber was not at all critical; it was varied in steps of 0.18 cm equivalent stopping power of air (0.13 Mev for protons).

Observations were made at voltages differing by 12 kv from 0.3 to 1.6 Mv. A single measurement required on the average about two minutes. The target was usually bombarded with 66 microcoulombs, corresponding to 10 divisions of the current integrator. The length of the run was varied, depending on the intensities under observation. Below 0.6 Mv the molecular ion beam was used.

At resonances, the deflections of the radiation recording electroscopes were of the order of 20 divisions. Background corrections, of the order of 0.5 division, depending on the length of the run, were applied to the electroscope readings. This correction was for the natural leakage, and did not include the effect of stray radiation excited in the tube. Between resonances the observed deflections were sometimes equal to the background correction. Judging from the reproducibility of the observations, the reliability of the electroscope data is about what one would expect from the fact that they can be read to about 0.1 division. The counting rate of the alpha-particles was sufficiently high that no background correction was needed, but not high enough to introduce errors through inability of the counter to resolve successive counts.

### The relative sensitivities of the electroscopes to gamma-radiation

The pair excitation was determined by subtracting from the total reading of the unshielded electroscope the effect of the gamma-rays as determined from the shielded electroscope. Thus it was necessary to know their relative sensitivities to gamma-rays in the positions in which the measurements were made. The principal part of the ionization associated with the gamma-radiation is produced by the scattered electrons and the electron pairs ejected from the surrounding matter. However, an important part is produced indirectly through the effects of scattered radiation and the radiation accompanying the annihilation of the electron pair positions. The magnitude of these secondary effects is greatly dependent upon the composition and geometrical arrangement of the material near the electroscope, especially when the material has a high atomic number, as in the present experiment. In view of the complicated nature of these effects and the difficulty of the calculations involved, it seemed best to make the comparison of the electroscopes without any change in the apparatus and using the gamma-radiation in question. This was possible, for rough absorption measurements showed that with a bombarding voltage of 0.335 Mev the intensity of pairs was very small. The presence of a few pairs would not modify the results appreciably. The sensitivity to gamma-radiation of the unshielded electroscope at the position used was found to be 2.0 times that of the shielded electroscope in its position. The pair yield was thus secured by subtracting twice the reading of the shielded electroscope from the reading of the unshielded electroscope.

### The absolute sensitivity of the electroscopes

To determine the absolute yield of gamma-rays or pairs a calibration of the absolute sensitivity of the electroscope employed must be made by noting its rate of deflection while exposed to radiation of known intensity. To facilitate calculations in such a measurement it is desirable to surround the electroscope with a medium so dense that within a distance equal to the range of the secondary electrons, the intensity and

composition of the radiation is uniform. One must be able to calculate the number of electrons ejected from this medium into the electroscope in terms of the radiation in question. Owing to the complications mentioned above and discussed in more detail below, this is not practicable if the instrument is surrounded by lead. However, if a material of low atomic number is used, these complicating effects become inappreciable and may be disregarded. It should be pointed out that lead is not objectionable as a surrounding medium in the determination of an excitation curve, where only varying intensities of a fixed kind of radiation are involved, but is objectionable when a measurement is to be made of the absolute intensities of one or more gamma-rays of different energy.

Let  $S$  be the number of ion pairs per cc produced per second at a distance of one cm from a radium source of one milligram under the conditions of the experiment. Then if  $D$  is the number of divisions per second recorded at a distance  $R$  from a source of  $M$  milligrams we have the reciprocal sensitivity of the electroscope given by:

$$c = SM/R^2D \text{ ion pairs per cc per div.}$$

In these experiments the electroscope was placed in the center of a paraffin sphere 6 inches in diameter and the deflection observed while it was exposed to the radiation from a 1.915-milligram radium standard in equilibrium with its decay products surrounded by 0.5 mm of brass and 1.0 mm of lead placed 100 cm from the electroscope.

The quantity  $S$  is given by  $sAB$  where  $s$  is the strength of a unit radium source with the filtration necessary to remove the very soft radium gamma-rays,  $A$  is a factor arising from absorption of the radiation in the walls of the electroscope, and  $B$  is the effectiveness, relative to air, of the medium surrounding the electroscope in applying secondary electrons to the electroscope.

Laurence<sup>24</sup> gives an empirical expression for the strength of a unit radium source filtered by platinum of thickness  $t$  in millimeters as follows:

$$s = 8.98(1 - 0.13t) \text{ roentgens/mg hr. per cm}^2.$$

<sup>24</sup> G. C. Laurence, Can. J. Research **A15**, 67 (1937).



This relation holds for  $t \geq 0.3$  mm. In our units

$$s = 5.19(1 - 0.13t) \\ \times 10^6 \text{ ion pairs per cc/mg sec. per cm}^2.$$

The platinum equivalent of the brass and lead filtration employed in these experiments was  $t = 0.66$  mm so that

$$s = 4.74 \times 10^6 \text{ ion pairs per cc/mg sec. per cm}^2.$$

With the softer components of the radiation removed, photoelectric absorption in the paraffin is unimportant. In view of the low atomic numbers of the constituents of paraffin, pair formation absorption may also be neglected. Data given by Lauritsen<sup>25</sup> show that the true absorption of energy by the Compton effect is roughly the same for all components of the radium radiation. The value  $\sigma_a = 0.9 \times 10^{-25}$  cm<sup>2</sup> for the electronic cross section<sup>25</sup> is a reasonable average value. The electron density in paraffin is  $n = 3.1 \times 10^{23}$  per cc so that the absorption coefficient and attenuation factor are, respectively,  $n\sigma_a = 0.028$  per cm and  $A = \exp(-n\sigma_a x) = 0.87$  for  $x = 5.0$  cm, the thickness of the paraffin effective in absorbing the radiation.

The effectiveness, relative to air, of the surrounding medium in supplying secondary electrons to the electroscopie must also be known. Laurence<sup>24</sup> designates this quantity by  $B$  and has computed it for those cases in which the Compton effect is the only important mechanism in the absorption of the radiation by the medium and the secondaries lose energy only through ionization by collision. For gamma-ray energies between 1 and 2 Mev the values of  $B$  for paraffin and for aluminum are constant to within 1 percent and are, respectively, 0.93 and 1.06. The walls of the electroscopie are of aluminum and have a thickness 0.038 cm which is about 20 percent of the range of the fastest electrons scattered by the radium radiation. Some average between the values for aluminum and paraffin must be taken. Since a large fraction of the gamma-rays have energies much less than the most energetic, and since the scattered electrons do not in general receive the maximum possible energy, it would seem reasonable to weight the two values equally, yielding the ratio  $B = 1.00$ .

<sup>25</sup> C. C. Lauritsen, Am. J. Roent. and Rad. Therapy **30**, 380, 529 (1933).

This combination of paraffin and aluminum is thus roughly equivalent to the ideal air equivalent walls.

We observed  $D = 0.062$  div. per sec. for the electroscopie used in the gamma-ray measurements so that for this electroscopie  $c = 1.27 \times 10^4$  ion pairs per cc per div. Its volume was 145 cc so that  $cV = 1.84 \times 10^6$  ion pairs per div. The corresponding quantities for the pair electroscopie were found to be  $D = 0.076$  div. per sec.,  $c = 1.04 \times 10^4$  ion pairs per cc per div.,  $V = 160$  cc and  $cV = 1.66 \times 10^6$  ion pairs per div.

#### The absolute yield of the gamma-radiation

The gamma-ray electroscopie, again with the lead shield removed, was placed inside the paraffin sphere and exposed to the radiation from a thick CaF<sub>2</sub> target, located just outside the sphere and bombarded with 1.04-Mev protons. This voltage was chosen because this point lies on a flat portion of the thick target excitation curve. The gamma-ray yield is given by  $Y_\gamma = (4\pi r^2 cd/IA) \times 1.6 \times 10^{-13}$  quanta per proton, where  $d$  is the number of divisions observed per microcoulomb of protons bombarding the target at a distance  $r$  from the electroscopie,  $I$  is the ionization per cc produced by a flux of 6.2-Mev radiation of one quantum per cm<sup>2</sup>, and  $A$  is the absorption factor for this radiation in the electroscopie walls. This equation assumes an isotropic distribution of the radiation, an assumption which is justified when the origin of the radiation is considered. A properly weighted value for  $r$  must of course be employed.

The essential point is now the calculation of  $I$ , which is in detail complicated in the high energy region where pair production, annihilation, and bremsstrahlen all contribute. We shall give here a treatment adequate for the cases where these effects are not too large, i.e., where the gamma-ray energy is under 25 Mev and the atomic number of the material surrounding the electroscopie is low. Since no nuclear radiation above 18 Mev has been observed, atomic screening which sets in at much higher energies will not be important.

For the region below 2 Mev several investigations<sup>24-26</sup> of  $I$  exist. The derivation depends

<sup>26</sup> L. H. Gray, Proc. Roy. Soc. **A156**, 578 (1936).

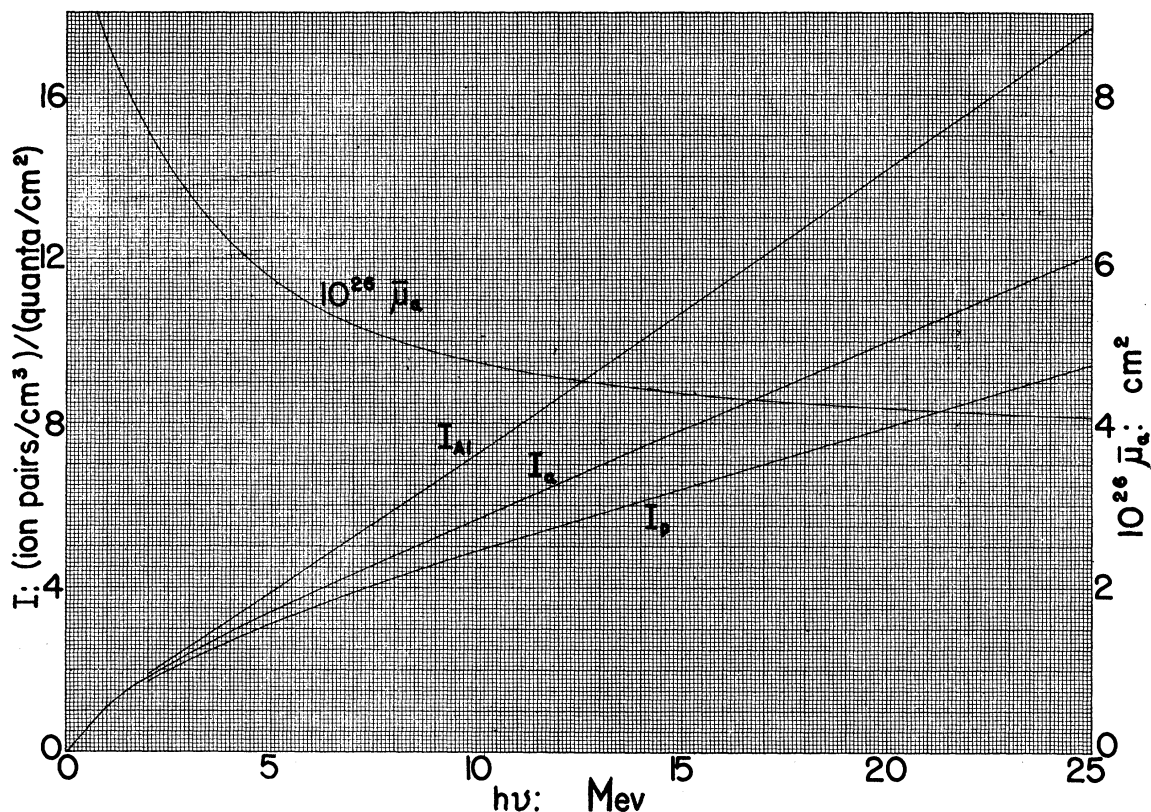


FIG. 2. The variation with energy of the quantities  $\mu_a$  and  $I$ . The quantity  $\mu_a$  is the true energy absorption cross section per electron for quanta of energy  $E$  in air. The quantity  $I$  is the number of ion pairs per unit volume produced by a flux of one quantum of energy  $E$  per square centimeter in an electroscop filled with air under standard conditions and having walls of aluminum (A), air equivalent material (a), and paraffin (p).

fundamentally on a theorem due to Gray<sup>26</sup> that the energy equivalent of the ionization measured per cc in a cavity in a solid medium is equal to the energy converted per cc in the medium ( $W_m$ ) divided by the relative stopping power ( $dE/dx$ ) of the medium and the gas filling the cavity for the secondaries produced by the radiation. Thus

$$\omega_g I = W_m \frac{(dE/dx)_g}{(dE/dx)_m},$$

where  $\omega_g$  is the energy lost per ion pair formed in the gas. This theorem assumes a cavity small compared to the range of the secondary electrons in the cavity, a uniform intensity and composition of radiation near the cavity, and that the relative stopping power is independent of the velocity of the secondaries. The first two conditions are satisfied in the energy range under discussion for electroscopes of a convenient size

(100 to 200 cc volume) when surrounded by a solid layer thicker than the maximum range of the secondaries as long as the source of radiation is not too close to the electroscop. In our experiments the source was probably too close to fulfill the second condition but we neglect this geometrical factor.

Laurence<sup>24</sup> has derived an analytical expression for Gray's theorem in such a way as to show that elastic scattering of the secondaries does not invalidate the theorem. Laurence also shows that if the relative stopping power is a function of the velocity of the secondaries it is only necessary to average it over the secondary energy. This is equivalent to averaging the stopping power in the gas over the path of the secondary in the medium as can be seen as follows:

$$\int_0^{E_0} \frac{(dE/dx)_g}{(dE/dx)_m} dE = \int_0^{R_0} \left( \frac{dE}{dx} \right)_g dx_m,$$

where  $E_0$  and  $R_0$  are the initial energy and full range of the secondary. In an extension to the energy range where radiation plays a part it is clear from the last expression that only the energy converted into ionization in the chamber must be used in computing  $(dE/dx)_g$  but that the full energy loss by radiation as well as ionization must be used in computing the range and hence  $(dE/dx)_m$  in the medium. Finally, averaging over the energy distribution of the secondaries, we can write Laurence's expression in the modified form:

$$I = \frac{1}{\omega_g} \int_0^{h\nu} \phi_m(h\nu, E_0) \int_0^{E_0} \frac{(dE/dx)_g'}{(dE/dx)_m} dE dE_0$$

or more conveniently,

$$I = \frac{N_g Z_g}{\omega_g} \int_0^{h\nu} \mu_m(h\nu, E_0) \int_0^{E_0} \frac{\lambda_g'}{\lambda_m} dE dE_0$$

×  $\frac{\text{ion pairs per cc}}{\text{quanta per cm}^2}$ ,

where  $I$  is the ionization per unit volume in the cavity per unit flux of radiation (quanta per  $\text{cm}^2$ ) in the medium,  $N_g Z_g$  is the number of electrons per cc of gas,  $\phi_m(h\nu, E_0) dE_0$  is the number of secondary electrons with energy between  $E_0$  and  $E_0 + dE_0$  produced per cm of path in the medium by a quantum of energy  $h\nu$ ,  $\mu_m(h\nu, E_0) dE_0$  is the corresponding electronic cross section for secondary production,  $(dE/dx)_g'$  is the stopping power by ionization of the gas,  $(dE/dx)_m$  is the total stopping power of the medium and  $\lambda_g'$  and  $\lambda_m$  are the corresponding electronic stopping cross sections. If the electroscopie is filled with air we have  $N_a Z_a / \omega_a = 1.20 \times 10^{19} \text{ (ev} \cdot \text{cc)}^{-1}$  using  $\omega_a = 32.5$  electron volts.

It will be seen that the complications involved in applying Gray's theorem are two in number.

1. The energy converted per cc in the medium is a complex function of the gamma-ray energy.

2. The relative stopping power of solid medium and gas is a complex function of their atomic number and of the energy of the secondaries. Of these two effects, the second can be almost completely eliminated by suitable choice of the surrounding medium (air equivalent) and can be made very large indeed by the use of Pb or other

heavy elements. In our work with paraffin and aluminum walls this second effect is quite small. For high energy quanta the complications in (1) cannot be eliminated, but can be reduced by using a medium of low  $Z$ .

We will first discuss (2). Using Bloch's formula for the stopping from ionization by collision and approximating to the stopping by radiation for the domain 2-25 Mev, by

$$\lambda_{\text{rad}}/\lambda = (\lambda - \lambda')/\lambda \approx EZ/2000 mc^2,$$

where  $E$  is the electron energy, we get

$$\frac{\lambda_a'}{\lambda_m} \approx 1 - \frac{EZ_m}{2000mc^2} + \frac{2 \ln(Z_m/Z_a)}{3 \ln(E/A)}$$

where

$$A = (m^3 ch R Z_m)^{1/2}.$$

This expression must be averaged, for each secondary, over all its energy and then over the initial energy distribution of the secondaries produced in the solid medium. The first average is

$$\left\langle \frac{\lambda_a'}{\lambda_m} \right\rangle_{Av} \approx 1 - \frac{E_0 Z_m}{4000mc^2} + \frac{2}{23} \ln \frac{Z_m}{Z_a},$$

where the factor  $2/23$  is a good average value in the domain under discussion.

To return to (1), we need to know  $\mu_m(h\nu, E_0) dE_0$ , the cross section for producing a secondary of energy  $E_0$  in the medium. If we set  $f = E_0/h\nu$  and  $\kappa = h\nu/mc^2$  the Klein-Nishina formula and the cross section for pair formation give at high energies:

$$\mu_m(\kappa, f) df = \pi r_0^2 df \left[ \frac{1}{\kappa} \left( 1 - f + \frac{1}{1 - f} \right) + 1.65 \frac{Z_m}{137} \frac{\kappa}{\kappa - 2} \ln \frac{\kappa}{4.3} \right],$$

where  $r_0 = e^2/mc^2$  and where the second term which is for pair production is an empirical fit to Heitler's curve<sup>27</sup> in the region from 2 to 25 Mev.

The remaining procedure is to integrate  $E_0 \langle \lambda_a' / \lambda_m \rangle_{Av}$  over the differential cross section. The results are plotted in Fig. 2 for paraffin ( $Z_m \approx 4.7$ ), air ( $Z_m = Z_a = 7.2$ ) and aluminum ( $Z_m = 13$ ) and are given in good numerical

<sup>27</sup> W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, 1935), p. 201.

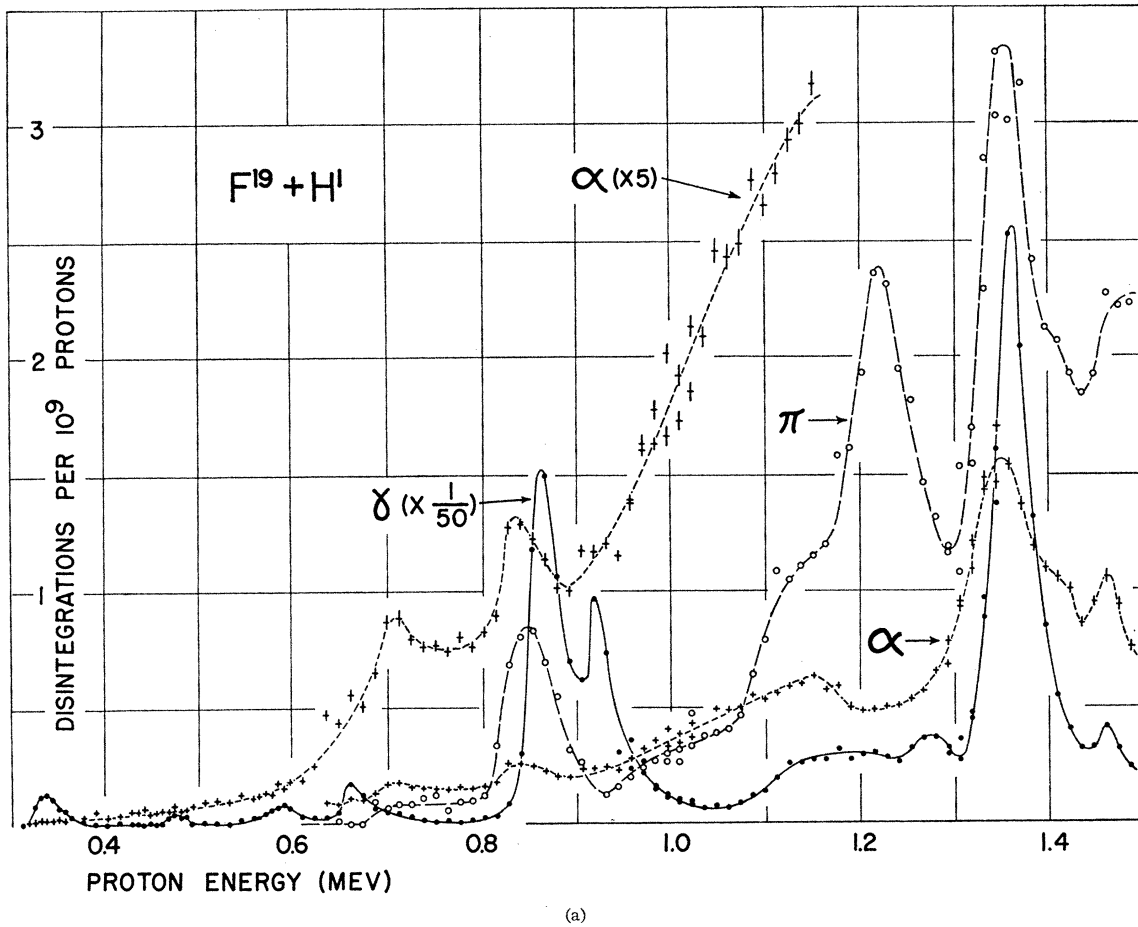


FIG. 3. The excitation functions. The curves marked  $\gamma$ ,  $\alpha$  and  $\pi$  refer to the gamma-rays, long range alpha-particles and electron pairs, respectively. In some regions Fig. 3(a) represents a combination of four independent sets of data; Fig. 3(b) (see opposite page) is based on a single set of data. The equivalent target thickness for 1-Mev protons was 8 kev for the upper curve, 1.4 kev for the lower curve. In Fig. 3(b) the upper end of the pair curve has been omitted; while its shape is not known accurately, it is certain that its sharp rise continues until at 1.62 Mev the intensity is about 50 percent greater than at 1.5 Mev.

approximation for  $Z_m < 20$  and  $h\nu < 25$  Mev by

$$I = (N_a Z_a / \omega_a) h\nu \bar{\mu}_m$$

ion pairs per cc/quanta per cm<sup>2</sup>,

where  $\bar{\mu}_m$  is the true energy absorption coefficient per electron for the medium as measured in an air cavity and is given by

$$\bar{\mu}_m = \pi r_0^2 \frac{mc^2}{h\nu} \left( 1 - \frac{h\nu Z_m}{5000mc^2} + \frac{2}{23} \ln \frac{Z_m}{Z_a} \right) \times \left( \ln \frac{h\nu}{1.1mc^2} + \frac{(h\nu - 2mc^2)Z_m}{165mc^2} \ln \frac{h\nu}{4.3mc^2} \right).$$

The absorption coefficient for air equivalent

walls ( $\bar{\mu}_a$ ) is also given in Fig. 2. The terms  $\pi r_0^2 mc^2 N_a Z_a / \omega_a = 1.51$  in our units.

For  $Z_m = 20$  and  $h\nu = 25$  Mev the expression for  $I$  is correct to about 5 percent. To extend this treatment to higher  $Z_m$  and  $h\nu$  would require giving up Gray's theorem, since then it is no longer possible to treat the range of the electrons as small, that of the gamma-rays as large, compared to the dimensions of the surrounding medium.

Experimentally it has been shown that for lead  $\bar{\mu}$  is only slightly greater than the minimum in the total cross section curve for radiation of both 6.2 Mev and 17.5 Mev.<sup>9</sup> Thus  $I$  for a lead lined chamber is proportional to the quantum

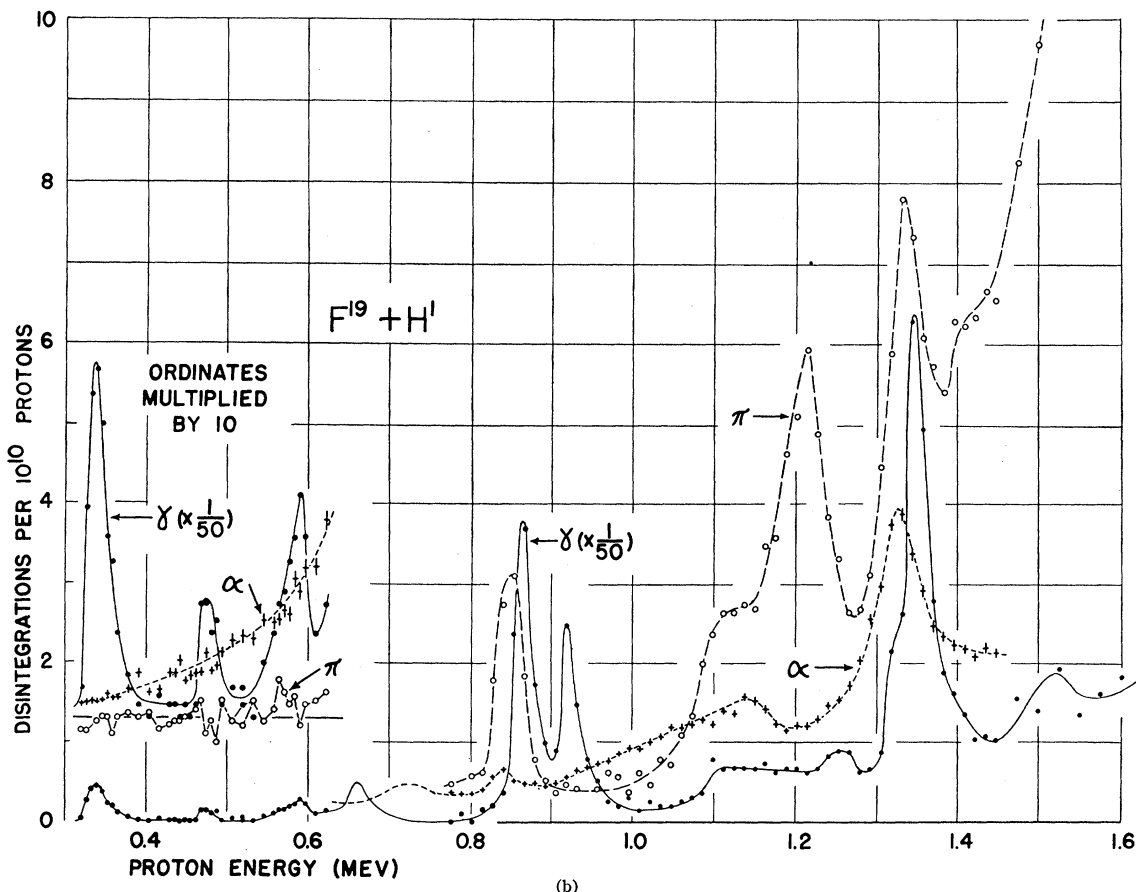


FIG. 3. (Continued).

energy being given very approximately by

$$I(\text{Pb}) = 0.9h\nu/mc^2$$

ion pairs per cc/quanta per  $\text{cm}^2$ .

The derivation for  $I$  given above permits of a calculation of the ions produced per unit volume of the electroscopie only when the energy and composition of the radiation in the wall is strictly known. In actual practice we wish to know the ionization produced by a monochromatic gamma-ray falling on the external electroscopie walls and some question may arise as to the complications arising from the building up of secondary radiation from the Compton effect, radiation of the secondary electrons, and annihilation of positron members of pairs. The answer to this question depends on the geometry of the experiment and only in case the secondary quanta produced in the electroscopie walls and escaping therefrom are completely compensated for by scattering in surrounding material can a

definite answer be given. Fortunately, almost complete compensation is attained by surrounding the electroscopie on all sides by walls thicker than the range of the secondary electrons in the wall material. A further simplification arises from the fact that  $\bar{\mu}$  is roughly independent of energy. If  $\phi(E)dE$  is the distribution in energy of quanta in the walls near the sensitive volume of the electroscopie produced by monochromatic quanta falling on the external electroscopie walls, then the energy conversion per unit volume is  $n \int \bar{\mu} E \phi(E) dE$  where  $n$  is the electron density in the walls. For  $\bar{\mu} = \text{constant}$  this becomes  $n \bar{\mu} W$  where  $W$  is the energy remaining in the form of radiation after the quanta have penetrated to a point where their secondaries can reach the electroscopie. Since secondaries are produced at all points in the wall surrounding the cavity we can write  $W = W_0 \exp[-n\bar{\mu}t]$  where  $t$  is the wall thickness. Actually  $\bar{\mu}$  is not strictly constant but we use the expression inserting the appropriate

value of  $\bar{\mu}$  for the incident radiation in question. For the 5-cm paraffin walls used in these experiments we find  $A = \exp[-n\bar{\mu}t] = 0.87$  for the filtered radium radiation and 0.92 for the  $F^{19} + H^1$  radiation.

The thick target yield was measured at one point (1.04 Mev) and the yield for each resonance was determined from the relative area under the thin target curve for that particular resonance. The results are discussed below.

### The absolute yield of the pair emission

The pair measurements were made with a minimum of material between the target and the electroscop. This material consisted of (1) the target backing = 0.010 inch of tantalum, (2) the target holder = 0.008 inch of phosphor bronze, (3) the target tube = 0.004 inch of German silver, and (4) the electroscop wall = 0.030 inch of aluminum. Both the target and the electroscop were surrounded by lead (Fig. 1). Under these conditions the yield of pairs can be computed from the expression

$$Y_{\pi} = \frac{4\pi r^2 c \omega_0 d \times 1.6 \times 10^{-13}}{n_0 \int \psi(E) \lambda_0' dE + n_0 \int \phi(E) \bar{\mu}_m E dE} \quad \text{pairs per proton,}$$

where  $r$ ,  $c$ ,  $\omega_0$ ,  $d$ ,  $\lambda_0'$ ,  $n_0$  and  $\bar{\mu}_m$  are analogous to quantities already defined for gamma-radiation,  $\psi(E)dE$  is the energy distribution per pair produced of pair members entering the electroscop and  $\phi(E)dE$  is the energy distribution per pair of annihilation quanta and bremsstrahlen quanta in the material surrounding the electroscop.

There is some uncertainty in the distribution in energy of the pair members although the cloud-chamber work of Fowler and Lauritsen<sup>21</sup> gave an energy distribution roughly constant up to the maximum kinetic energy  $E_M = 4.9$  Mev. If all the electrons lose the same amount of energy  $E_L$  or are stopped, if their initial energy is less than that amount, in passing through the material between the target and the ionization chamber, the distribution in energy of the electrons in the chamber will be approximately constant up to a maximum energy,  $E_M' = E_M - E_L$ . Then

$$\begin{aligned} n_0 \int \psi(E) \lambda_0' dE &= n_0 \bar{\lambda}_0' \int_0^{E_M'} (2/E_M) dE \\ &= 2E_M'^2 / E_M R_{M'}', \end{aligned}$$

where  $E_M'/R_{M}'$  is taken as a good average value for  $n_0 \lambda_0'$ ,  $R_{M}'$  being the range of an electron of energy  $E_M'$ , neglecting radiation.

The energy radiated by the pair members over their ranges in the lead box surrounding the electroscop and target can be computed by averaging  $EZ_m/2000mc^2$  over their energy distribution. We find

$$\bar{E}_{\text{rad}} = \frac{1}{3} \frac{E_M Z_m}{2000mc^2} E_M.$$

In these experiments  $Z_m = 82$ ,  $E_M = 4.9$  Mev,  $E_{\text{rad}} = 0.66$  Mev which is equivalent to 1.3 annihilation quanta. This number must be added to

TABLE I. Yields of the  $F^{19} + H^1$  reactions in disintegrations per proton from a thick  $CaF_2$  target. In successive columns are recorded the proton energies for particular resonances or voltage regions, the radiation observed, the yield per  $10^7$  protons, the estimated true widths of the resonance peaks in kev, the stopping cross section in  $10^{-15}$  ev·cm<sup>2</sup> of  $CaF_2$  per fluorine atom, the proton wave-lengths in  $10^{-12}$  cm, and values for  $\omega\gamma = \omega\Gamma_p\Gamma_z/\Gamma$  in ev. The proton energies for the  $\gamma$ -ray resonances are those given by Bernet, Herb and Parkinson. The value zero for a resonance width indicates a width which is probably very small and certainly less than 10 kev. The great width of several peaks may be due to the superposition of two or more resonances. The nonresonant yields for pairs and alphas are the integrated yields to 1.5 Mev below smooth curves drawn through the minima in the excitation functions for pairs and alphas.

$E_P$ (MEV)	R	$Y \times 10^7$	$\Gamma$ (KEV)	$\epsilon \times 10^{15}$ (EV·CM <sup>2</sup> )	$\lambda \times 10^{12}$ (CM)	$\omega\gamma$ (EV)
0.334	$\gamma$	0.18 <sub>5</sub>	0	24.1	5.20	33
0.479	$\gamma$	0.05 <sub>2</sub>	0	18.4	4.34	10
0.589	$\gamma$	0.24 <sub>2</sub>	25	15.5	3.92	49
0.660	$\gamma$	0.46 <sub>2</sub>	0	14.2	3.70	96
0.6-0.8	$\pi$	0.013 <sub>2</sub>	—	—	—	—
0.72	$\alpha$	0.007 <sub>5</sub>	15	13.6	3.54	1.6
0.84	$\alpha$	0.006 <sub>1</sub>	15	12.3	3.30	1.4
0.85	$\pi$	0.10 <sub>5</sub>	15	12.1	3.26	24
0.862	$\gamma$	3.34	0	12.0	3.24	760
0.927	$\gamma$	2.21	0	11.5	3.12	520
0.9-1.2	$\alpha$	0.06 <sub>4</sub>	—	—	—	—
1.14	$\pi$	0.07 <sub>3</sub>	30	10.0	2.81	20
1.22	$\pi$	0.20 <sub>5</sub>	30	9.5	2.72	53
1.1-1.3	$\gamma$	3.03	—	—	—	—
1.35	$\alpha$	0.13 <sub>5</sub>	25	8.8	2.59	36
1.35	$\pi$	0.19 <sub>5</sub>	25	8.8	2.59	51
1.335	$\gamma$	1.25	0	8.9	2.60	330
1.363	$\gamma$	7.71	10	8.7	2.57	2020
Nonresonant	$\pi$	1.18	—	—	—	—
	$\alpha$	0.69	—	—	—	—
Total						
to 1.0	$\gamma$	6.50	—	—	—	—
to 1.5	$\gamma$	22.0	—	—	—	—
"	$\pi$	1.78	—	—	—	—
"	$\alpha$	0.90	—	—	—	—

the two quanta resulting from the annihilation of the positron member of the pair.

The effective secondaries from these quanta were produced in the aluminum walls of the electroscopes and inserting the proper value for  $\bar{\mu}_m$  we find that the radiation term contributes 2 percent of the denominator of the expression for  $Y_\pi$ . The quantity  $E_L$  was found to be 1.3 Mev and the range of an electron of energy  $E_M' = 3.6$  Mev was taken to be 14 meters in air. Hence  $2E_M'^2/E_M R_M' = 3770$  ev per cm.

### The yield of the long range alpha-particles

In the case of the alpha-particles the assumption of isotropic distribution is not justified. Their angular distribution has been measured by Ellett, McLean, Young and Plain<sup>28</sup> at voltages from 0.27 to 0.44 Mev. The distribution is practically independent of the proton energy in this range and is described by

$$I(\theta) = 1 + 0.77 \cos \theta + 0.17 \cos^2 \theta$$

in the center of mass coordinates. Lacking knowledge of the distribution at higher energies all of the calculations of yield from the alpha-particle intensity perpendicular to the proton beam were made on the basis of an isotropic distribution in laboratory coordinates. If instead, the calculations were based on the above formula, the results would be increased by from 6 percent, at the lower voltage, to 10 percent at the higher voltages. The calculation is made from obvious geometrical considerations.

## EXPERIMENTAL RESULTS

### The excitation functions and absolute yields

In Fig. 3 are shown the excitation curves obtained on two occasions. The ordinates are given in terms of the number of transmutations per incident proton. For thin targets with a stopping power less than the fluctuations in the tube voltage the ordinates will be proportional to the target thickness. For the second curve a thinner target was used. This accounts for the lower intensity and better resolution. It should be emphasized that the radiation designated as pairs is just that soft component absorbed by  $\frac{1}{8}$

inch of lead; in this experiment no further attempt was made to establish the identity of the radiation. As remarked above, the nature of this radiation has been established definitely only at 0.82- and 1.13-Mev bombarding energy.

As stated above, no correction has been applied for undesired radiation from the tube itself, but some control runs were made with a clean tantalum target. The measurements are not very reproducible, but they show that the stray radiation gives a significant background only in the neighborhood of 1.05 Mev where the true gamma-ray intensity is much smaller than shown, and in the region from 1.1 to 1.3 Mev where the gamma-ray intensity is about 25 percent less than that plotted.

It will be observed that the gamma-ray curves are in good agreement with the work of Bernet *et al.*, and the other investigators although the resolution is not quite as good as has been obtained before.

In Table I are given the absolute yields for all resonances below 1.5 Mev. As may be seen by a comparison with Fig. 3, some of the peaks tabulated are superimposed on a continuous background which makes an estimate of the yield to be attributed to the resonance somewhat difficult. In constructing the table, data on gamma-rays given by Bernet *et al.* have been used to fix the energy scale. The full widths at half-maxima for the various resonances were found by subtracting from the observed widths the width due to fluctuations in the tube voltage (25 kev). When zero is given as the width it must be interpreted as an indeterminate width certainly less than 10 kev and probably much smaller.

The yield of a nuclear reaction initiated by proton bombardment is given by

$$Y_X = \frac{\lambda^2}{2\epsilon} \omega \gamma = \frac{\lambda^2}{2\epsilon} \frac{\Gamma_P \Gamma_X}{\Gamma}$$

where the  $\Gamma$ 's are the appropriate decay constants in energy units,  $\lambda$  is the wave-length of the incident protons,  $\omega$  is a statistical weight factor which probably ranges in value from  $\frac{1}{4}$  to  $\frac{7}{4}$  and  $\epsilon$  is the energy loss cross section of the incident particle in the target material. The yield is thus a measure of the quantity  $\omega \gamma$ , which is listed in

<sup>28</sup> Ellett, McLean, Young and Plain, Phys. Rev. 57, 1083(A) (1940).

Table I for each resonance. We also include  $\lambda$  and  $\epsilon$  for each resonance. The stopping power of  $\text{CaF}_2$  relative to air was taken as 2.0 independent of the proton velocity and the variation of  $\epsilon_{\text{air}}$  was taken from a curve given by Bethe.<sup>29</sup>

From data given by Tuve and Hafstad,<sup>30,15</sup> Bethe<sup>31</sup> has estimated the yield of gamma-rays from the 0.334-Mev resonance with a  $\text{CaF}_2$  target. The result is about 200 times smaller than that obtained in the present experiment. The calculation depends on the cross section at the 0.440-Mev resonance in the production of gamma-rays by proton bombardment of lithium. For this quantity Tuve and Hafstad estimate  $10^{-27}$  cm<sup>2</sup>. They do not give the details of the calculation but indicate that it is not very reliable. Their observations were also made with a Lauritsen electroscope. Although the electroscope readings taken by Tuve and Hafstad near the 334 resonance appear to be only somewhat smaller than we would calculate from ours when account is taken of the Pb electroscope walls used in their work, the cross section which they give and thus Bethe's estimated yield is smaller than ours by a factor of 200 and seems to be seriously in error. Recent measurements in the same laboratory by Van Allen and Smith<sup>30</sup> on the yield of the short range alphas from the 334 resonance give a value of  $8.9 \pm 0.5 \times 10^4$  alphas per microcoulomb or  $1.43 \pm 0.8 \times 10^{-8}$  alpha per proton in excellent agreement with our yield of  $1.8 \times 10^{-8}$  gamma-ray per proton.

### Competitive resonances

The importance of establishing definitely any coincidence in the energy of resonances in different modes of disintegration of the intermediate nucleus has been previously discussed. Such a coincidence indicates that the disintegrations can be considered as competitive modes of decay of the same state of the compound nucleus.

In searching for coincidences in the energy of resonances in different processes it must not be overlooked that there is some possibility for an apparent coincidence between two different kinds

of resonances which do not involve the same state of the intermediate nucleus. For example, if  $N_\alpha$  alpha-particle resonances,  $N_\gamma$  gamma-ray resonances and  $N_\pi$  pair resonances are distributed at random over an energy range  $W$ , the number of pairs of different kinds of resonances separated in energy by less than  $\omega$ , is on the average:

$$n_{\alpha\gamma} = 2(\omega/W)N_\alpha N_\gamma$$

and so forth. In the present case  $N_\alpha=4$ ,  $N_\gamma=8$ ,  $N_\pi=4$  so that if we take  $\omega=10$  kev we obtain  $n_{\alpha\gamma}=0.43$ ,  $n_{\alpha\pi}=0.21$  and  $n_{\gamma\pi}=0.43$ .

We actually find two coincidences within 10 kev; namely, those between pairs and long range alphas at 0.85 Mev, and 1.35 Mev. On the basis of the above calculations it seems difficult to attribute both of these to accidental coincidences. On the other hand, a close comparison of the curves suggests that in both cases the peaks are slightly separated. A statistical study to determine the reality of the separation was made as follows: On a plot of the observations of each kind of radiation a reasonable curve was drawn to represent the background upon which the resonance is superimposed; this background intensity function was then subtracted from the observed intensity, and the centroid of the remaining intensity function was taken as the resonance energy. The separation at 0.85 Mev was found to be  $9 \pm 5$  kev; the uncertainty is the probable error. This must be compared with the true widths of the peaks which we have estimated as 15 kev. The very prominent pair and long range alpha-particle peaks near 1.35 Mev appear to be coincident. A statistical investigation gives for the separation  $6.5 \pm 1.5$  kev while the widths were both estimated to be 25 kev.

In estimating the probable error in the separation no account was taken of the fact that the observations were made at 12-kev intervals. We think it likely, however, that these separations are real. In both cases the long range alpha lies below the pair resonance in the direction to be expected but by an amount very much larger than that to be expected from the differential variation of the probabilities for alpha-emission over the line breadth. It has been suggested by Professor J. R. Oppenheimer that the observed dissimilarity in long range alpha and pair backgrounds must bring with it differences in the

<sup>29</sup> H. A. Bethe, Rev. Mod. Phys. 9, 270 (1937).

<sup>30</sup> L. R. Hafstad and M. A. Tuve, Phys. Rev. 48, 306 (1935); Van Allen and Smith, Phys. Rev. 59, 108(A) (1940).

<sup>31</sup> H. A. Bethe, Rev. Mod. Phys. 9, 69 (1937).



interference effects between resonance and background, at least in certain angular ranges, sufficient to give the apparent displacement of the peaks. Since the pair background is relatively the smaller, the displacement from resonance will be mainly in the long range alpha-curve and the resonance energies in Ne<sup>20</sup> are best taken from the pair curve.

There are several gamma-ray resonances which are within 20 kev of pair resonances but both displacements and line breadth differences are here much too great to be accounted for by interference with the small backgrounds for pairs and gamma-rays.

DISCUSSION

The character of the states

An energy level diagram is shown in Fig. 4. The relative positions of the ground states are determined from the masses. The levels <sup>γ</sup>O<sup>16</sup> and <sup>π</sup>O<sup>16</sup> are plotted from a knowledge of the gamma-ray and pair energies. The first six excited states in neon are those found by Bonner. The higher levels are those found in the present and similar experiments.

If we accept the evidence discussed above as indicating competitive resonances in the production of pairs and long range alpha-particles, it is simplest to interpret this as a branching process in the decay of certain states of Ne<sup>20</sup>. According to Oppenheimer and Schwinger<sup>22</sup> this is reasonable; at energies above 1 Mev the effect of the Coulomb barrier is unimportant for both the long and short range alpha-particles. Following this interpretation the excited states of Ne<sup>20</sup> produced by proton bombardment of fluorine were classified in four groups, according to the manner of their decay, as indicated in the diagram. As mentioned before, this conclusion implies that the parity of the state <sup>π</sup>O<sup>16</sup> is even, and that it is possible to account for the pair emission through electromagnetic forces.

To explain the appearance of short range alpha-particles leading to gamma-emission rather than the long range alpha-particles we assume that the states <sup>γ</sup>Ne<sup>20</sup> and <sup>γ</sup>O<sup>16</sup> have odd parity and even angular momentum, or vice versa. Then the emission of both the short range alpha-particle leading to pairs and the long range alpha-particle

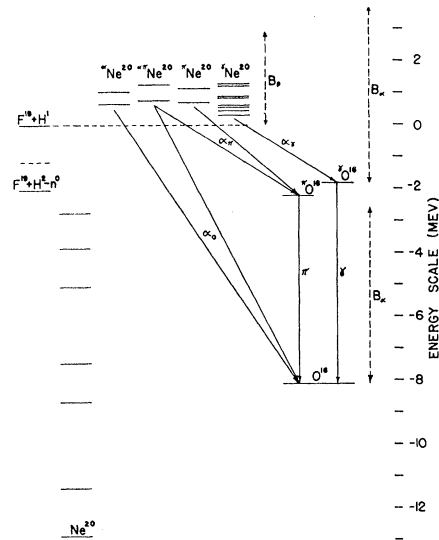


FIG. 4. Energy level diagram. The levels of Ne<sup>20</sup> above the level F<sup>19</sup>+H<sup>1</sup> are those observed in this and similar experiments. The levels of Ne<sup>20</sup> plotted in the column above the ground state of Ne<sup>20</sup> are those found by Bonner. The short dotted line in the left-hand column indicates his bombarding energy; the region between here and the level F<sup>19</sup>+H<sup>1</sup> is unexplored. The transitions marked α<sub>0</sub> are described in reaction (1), those marked α<sub>γ</sub> and γ in reaction (5), and those marked α<sub>π</sub> and π in reaction (7). The arrows marked B<sub>p</sub> and B<sub>α</sub> show the barrier heights for protons and alpha-particles, respectively.

are forbidden. The simplest assumption is that some of the states of <sup>γ</sup>Ne<sup>20</sup> are of the type (1, +) (angular momentum 1, and even parity) and that some of the states <sup>π</sup>Ne and <sup>απ</sup>Ne are of the type (0, +). These two different kinds of states of the neon nucleus might be formed by the two ways of adding the angular momenta of the fluorine nucleus and an s proton. The <sup>α</sup>Ne states can be assumed to have even parity but probably have even angular momenta greater than zero.

Remaining difficulties

However the picture is far from complete. No explanation has been advanced for the irregular variation in intensity of the various gamma-ray levels. More puzzling still is the very high probability of gamma-ray production compared with the other two processes. All three processes depend on the emission of an alpha-particle from the neon nucleus, and the effect of the barrier favors the long range alpha-particles. This is however compensated for in part by the higher probability of an alpha-particle having an energy

near the average rather than the total excitation energy in the compound nucleus. The three types of states  ${}^{\alpha}\text{Ne}^{20}$ ,  ${}^{\pi}\text{Ne}^{20}$  and  ${}^{\alpha\pi}\text{Ne}^{20}$  are differentiated on the basis of the relative intensities of the long and short range alpha-particles which result from their decay, but a complete explanation for this difference in behavior has not been proposed. Of course the irregular variation of the ratios of the yields of long range alphas to pair-alphas may be understood in part in terms of the fact that large changes in angular momentum would decrease the yield of the shorter range pair-alphas. But the consistently large ratios of gamma-alphas to pair-alphas can apparently not be understood on this basis, especially since  ${}^{\gamma}\text{Ne}^{20}$  and  ${}^{\pi}\text{Ne}^{20}$  can both

be formed by  $s$  collisions and both  ${}^{\gamma}\text{O}^{16}$  and  ${}^{\pi}\text{O}^{16}$  by the ejection of an  $s$  alpha-particle. It is possible that a new selection rule is involved.<sup>32</sup>

The yield measurements must be supplemented by precise measurements of the total decay width  $\Gamma$  of each resonance and of the distribution in angle of the alpha-particles before a complete description of the properties of the levels of the intermediate nucleus can be given.

In conclusion we wish to express our appreciation to Professor J. R. Oppenheimer for numerous contributions to the theoretical aspects of this discussion.

<sup>32</sup> Streib, Fowler and Lauritsen, Phys. Rev. **58**, 187(A) (1940).

## Velocity-Range Relation for Fission Fragments

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Considerations indicated in an earlier note as regards the rate of velocity loss of fission fragments along the range are developed in greater detail and a comparison is given between the calculations and more recent experiments. Especially is a more precise estimate given for the charge effective in electronic encounters which are determining for the stopping effect over the first part of range, and for the screening distance in nuclear collisions which are responsible for the ultimate stopping. In the estimate of the effect of electronic interactions, use is made of a comparison with the stopping of  $\alpha$ -particles of the same velocities. In this connection, however, a certain correction is necessary due to an intrinsic difference in the stopping formulae to be applied in the two cases. Moreover, fission fragment tracks show, in contrast to  $\alpha$ -rays, a considerable range straggling originating in the end part of the range. It is shown that in this respect also the calculation agrees closely with the experimental data.

**I**N an earlier note<sup>1</sup> the peculiar velocity-range relation for fission fragments revealed by cloud-chamber studies of fragment tracks<sup>2</sup> has been briefly discussed. In particular, it was pointed out that in the different parts of the range we have to do with two essentially different stopping mechanisms. At the beginning of the range, where the total charge of the fragment is still large, the stopping is due practically only to energy transfer to the individual electrons in the atoms of the gas penetrated. With decreasing

velocity, however, the fragment charge effective in electronic interactions will rapidly decrease and direct transfer of momentum from the fragment to the gas atoms through close nuclear collisions will gradually become of greater importance. In the last part of the range, such collisions will, in fact, be almost entirely responsible for the stopping effect. In the note it was shown that it is possible, from very simple considerations regarding the way in which the charge of the fragment varies with velocity, to account at least qualitatively for the characteristic features of the experimental velocity-range relation. The continuation of the work,

<sup>1</sup> N. Bohr, Phys. Rev. **58**, 654 (1940).

<sup>2</sup> K. J. Broström, J. K. Bøggild and T. Lauritsen, Phys. Rev. **58**, 651 (1940).