general principle outlined above, rather than as an adequate separation of cobalt-ion from the complex salt.

The experimental results are outlined in Table I. It should be noted that these ratios have not been "corrected." That is to say, they are ratios peculiar to the particular quantities of complex compound and "carrier" metal chloride used in these experiments. These ratios can be increased many-fold (a) by increasing the quantity of complex compound, and/or (b) reducing the quantity of "carrier" metal chloride used. In all these cases, therefore, the method is very successful in concentrating virtually all the radioactivity.

CONCLUSION

The general principle therefore appears to be established that a metal complex compound whose optically active isomers do not racemize can be used to concentrate radioactivity produced by simple neutron capture. Several points should again be noted in this connection. It is not necessary to resolve such a compound into its enantiomorphs in order to concentrate the metallic radioactivity. The racemic compound will, of course, be suitable. Secondly, the above criterion is a sufficient one, but not a necessary one. Other criteria undoubtedly will serve as well. It may well be that even those complex compounds whose enantiomorphs racemize rapidly can still be used.

TABLE I. *Ratio of concentrate activity to activity of the complex salt.*

Element	Rh	Ir	Pt	Co	Ru
Activity ratio	150	56	44	10	?*

* No activity detectable in either chemical fraction.

The support of the Research Corporation is gratefully acknowledged. The cooperation of the American Platinum Works of Newark, New Jersey, which supplied the iridium and ruthenium chlorides, is much appreciated.

MARCH 15, 1941 PHYSICAL REVIEW VOLUME 59

The Absolute Number of Quanta from the Bombardment of Fluorine with Protons†

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(Received January 30, 1941)

From recent experiments it seems very probable that the 6.2-Mev quanta from the proton bombardment of fluorine are associated in one-to-one correspondence with the short range alpha-particles. This correspondence makes possible the establishment of the gamma-ray intensity by measuring the alpha-particle yield. With a specially designed variable pressure absorption-cell ionization-chamber to facilitate clear distinction between the short range alpha-particles and the scattered protons, the angular distribution and total yield of the alpha-particles at the lowest resonance (330 kv) have been determined. The determinations were based on plateaus appearing in curves of counting rate as a function of (1) cell pressure, (2) counter bias, and (3) bombarding voltage at angles of 60°, 90°, 120°, and 150° with respect to the proton beam. The results show that: (*a*) To within two percent the angular distribution of the alpha-particles is spherically symmetric; and (*b*) the yield over 4π steradians is $8.9 \pm 0.5 \times 10^4$ alpha-particles per microcoulomb of 360-kv protons bombarding a thick crystal of CaF_2. The absolute number of quanta from the reaction is presumably the same to the same accuracy. With both a heavily shielded counter and a similarly protected electroscope the ratio of the gamma-ray intensity at 1050 kv to that at 370 kv was found to be 42.0 ± 0.8. Thus the gamma-ray intensity at 1050-kv bombarding voltage is $3.7_4 \pm 0.2 \times 10^6$ quanta per microcoulomb of protons on CaF_2. The angular distribution of the gamma-radiation was found to be spherically symmetric to within the experimental error of five percent. Knowledge of the absolute intensity of the fluorine source greatly enhances its value in photo-nuclear and other experiments.

INTRODUCTION

IN connection with experiments on photo-nuclear effects a source of high energy gamma-radiation of known quantum-intensity is needed; monochromatic radiation also is very desirable. The best such natural source is ThC″ ($E_\gamma = 2.65$ Mev). Of the artificial sources which have been investigated the resonance reactions of protons

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† Reported at November, 1940, meeting of American Physical Society, Chicago, Illinois.

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on fluorine ($E_\gamma = 6.2$ Mev) and protons on lithium ($E_\gamma = 17.5$ Mev) are the ones which yield gamma-rays best suited to quantitative measurements.

The mechanism of the fluorine reaction suggests a method of quantum calibration which is ideal in its simplicity and directness. There now exists strong evidence[1] to show that the disintegration proceeds as follows:

$$F^{19}+H^1 \rightarrow O^{16*}+He^4+Q_1, \qquad (a)$$

$$O^{16*} \rightarrow O^{16}+\gamma+Q_2, \qquad (b)$$

where $Q_1 = 1.79$ Mev and $Q_2 = 6.2$ Mev. The evidence consists of the facts (1) that the alpha-particle resonances occur within the experimental error at the same bombarding voltage as the gamma-ray resonances, (2) that the alpha-particle in (*a*) takes up the expected fraction of the energy of the incident proton while the gamma-ray has the same energy at all resonances investigated,[2] that is, that Q_1 and Q_2 are constants, and (3) that $Q_1+Q_2=Q_3$, where Q_3 ($=7.95$ Mev) is the energy released in the production of long range alpha-particles:[3]

$$F^{19}+H^1 \rightarrow O^{16}+He^4+Q_3. \qquad (c)$$

The one-to-one correspondence[4] in the emission of an alpha-particle and a quantum according to (*a*) makes possible the establishment of the gamma-ray intensity by measuring the alpha-particle yield.

The angular distribution and absolute yield of these heavy particles have been measured; in view of the evidence outlined above, this yield is presumably equal to the quantum intensity of the gamma-radiation.

The 330-kv resonance was chosen for two reasons: (1) It is probable that no pairs are emitted at this resonance;[5] (2) it is possible only at this resonance for reasons stated below to detect the alpha-particles by range differentiation.

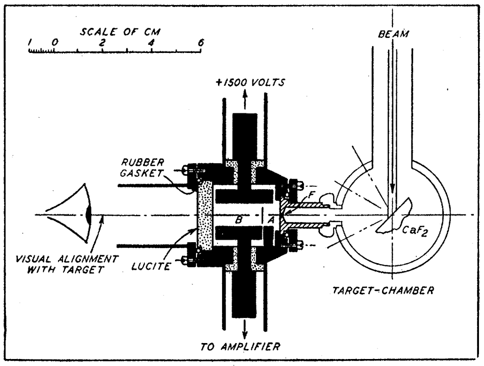

FIG. 1. The variable pressure absorption-ionization cell. The method of visual alignment is indicated in the drawing. (Details of beam collimation are given in Fig. 5.)

Once the absolute intensity at 330 kv is established, then a valid intensity comparison with the higher resonances can be made by a counter or ionization chamber, since the character of the radiation is the same.[2]

EXPERIMENTAL

The absorption cell

Even at 330 kv the difference between the ranges of the alpha-particles and the protons scattered elastically from the target is small and the scattered protons far exceed in number the disintegration particles. Under practical counting conditions the region in which the two types of particles can be clearly distinguished is less than one millimeter of range. Thus, a counting method of high resolution is required. For the final measurements a variable pressure absorption-ionization cell of the type used by Allison and associates[6] was designed so as to be well adapted to yield measurements (Fig. 1). It is to be noted that in this chamber the absorption cell and the ionization chamber operate at the

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[1] W. B. McLean, R. A. Becker, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **55**, 796 (1939); W. E. Burcham and C. L. Smith, Nature **143**, 795–796 (1939); W. E. Burcham and S. Devons, Proc. Roy. Soc. **A173**, 555–568 (1939).

[2] P. I. Dee, S. C. Curran and J. E. Strothers, Nature **143**, 759–760 (1939); and L. A. Delsasso, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **51**, 527 (1937).

[3] M. C. Henderson, M. S. Livingston and E. O. Lawrence, Phys. Rev. **46**, 38–42 (1934).

[4] Above 0.68 Mev, Lauritsen and others have shown that another excited state of the oxygen nucleus is associated with a short range alpha-particle and can lose its energy by electron-pair emission according to $O^{16**} \rightarrow O^{16}+\pi+5.9$ Mev. See W. A. Fowler and C. C. Lauritsen, Phys. Rev. **56**, 840–841 (1939).

[5] Private communication. Streib, Fowler and Lauritsen state that at this resonance their method of detecting nuclear pairs would have revealed as few pairs as one-half percent of the number of quanta observed.

[6] S. K. Allison, L. C. Miller, G. J. Perlow, L. S. Skaggs and N. M. Smith, Jr., Phys. Rev. **58**, 178 (1940); G. J. Perlow, Phys. Rev. **58**, 218–225 (1940); and L. C. Miller, Phys. Rev. **58**, 935–942 (1940). See H. Geiger, Zeits. f. Physik **8**, 45–57 (1921) for an early use of the usual form of absorption cell.

same pressure, which can be varied widely. In the ordinary case the pressure is varied in only the absorption cell and the ionization chamber works at atmospheric pressure. However, the two methods have similar characteristics[6] as long as the ionization chamber in the former case is deep, that is, as long as the particle is completely stopped by the air in the sensitive region. If the plates of the ionization chamber are perpendicular to the path of the particle, two disadvantages result: In order to have a deep chamber an inconveniently high sweeping voltage is required, and a metallic screen or foil is needed to separate the absorption region from the collection region. Screens reduce the effective aperture in an uncertain manner. Foils introduce undesirable stopping and, if quite thin, are subject to acoustical disturbances.

The use of plates parallel to the average path of the alpha-particles rather than perpendicular makes it possible to separately define the absorption region *A* and the ionization chamber *B* without the use of screens or foils. This is accomplished by the fact that the lines of force are perpendicular to the path of the particle and all the positive ionization in the region *A* is attracted to the grounded electrode, all in *B* to the grid electrode of the amplifier. Furthermore, one can make the sensitive region very deep—in this case 17 mm—increasing the useful pressure range of the instrument without requiring an unusually high voltage (1500 volts was used on the plates for the spacing of 8 mm). The range dispersion was made high by constructing the absorption region *A* only 7 mm deep. Thus a change of pressure of 10 cm of mercury in the cell corresponded to a change in stopping power of roughly 1 mm of normal air. This is indicated by the horizontal arrow in Fig. 2. This design reduces the instrumental straggling (and consequently increases the resolution) by affording complete electrical and acoustical shielding. The first stage of the amplifier was isolated mechanically from the cell.

The aperture *F* was the only one which limited the solid angle subtended from any point of the effective target. This arrangement simplified the calculation of the total yield. The plates of the cell were sufficiently far apart (8 mm) so that alpha-particles leaving anywhere from a target area 9 mm in diameter would enter the cell and be detected. The maximum size of the beam at the target was a circle of only 4 mm diameter. In addition to this precaution the "Lucite" window on the back of the cell permitted its accurate alignment by viewing through the window and through the aperture *F* the blue fluorescent spot produced by the proton beam on the calcium fluoride crystal used as a target. The cell was adjusted until the light from the average position of the fluorescence passed through the center of the chamber and was safely distant from the collection plates. In this manner it was assured that all particles coming from the target and penetrating the window *F* would be counted. A brass shielding cap was put over the "Lucite" window while it was not being used to view the target-cell alignment.

Beam collimation and measurement

The beam of protons was collimated by two holes (*A* and *B* in Fig. 5) 0.32 cm in diameter and 22 cm apart. The target was located 16 cm below *B*. Therefore, the maximum possible excursion of the beam at the target was within a circle of 4 mm diameter. Measurements of the carbonized spot on the surface of the target after bombardment proved this to be the case. The electrode *C* was maintained at -45 volts to prevent the loss of secondary electrons from the target chamber. The electrode *D* was grounded to prevent leakage from *C* to the target. No change in the beam current reading (sensitivity one percent) could be observed for a variation of the potential of *C* from 0.0 to -135 volts. No leakage in the order of 0.001 microampere could be observed between *D* and the target-chamber when 135 volts were applied. Secondary electron-current to the grounded window frame *F* was not measurable and was therefore less than 0.1 percent. A slight charging up of the target-face was probably helpful in preventing secondary electrons from leaving the target. The current to the target was read at 10-second intervals during the runs with a Sensitive Research model 55 microammeter and the average taken. This meter was calibrated in two ways by comparing it (1) with a Leeds and Northrup standard resistor and Weston laboratory standard voltmeter and (2) with a previously unused Sensitive Research model J–W microammeter. The two methods gave the same result to within less than one-half percent. The bombarding current was held low to values of the order of 3 microamperes to prevent superposition of the pulses caused by the scattered protons. All runs during which the current fluctuated seriously were discarded.

It is unlikely that the average value of the current over a set of runs was in error by more than one percent. The magnetically separated proton beam was used in all measurements. The fact that no measurable current was ever found between the mass-one and mass-two spots made it certain that no appreciable number of molecular ions of partial energy was present in the mass-one spot. The current in the single mass spot usually constituted about 80 percent of the total beam down the accelerating tube.

Thin-window calibration

The aperture F was a single hole of about 0.05 cm diameter made vacuum tight by a collodion film (pharmacist's "Newskin") of 1 to 2 mm air equivalent. The film was prepared in the usual way by spreading of the collodion solution on water and sealed to the window frame with "Apiezon W." Examination of the films with a low power microscope usually showed the presence of a slight amount of dust. The presence of these foreign particles had the effect of decreasing the effective aperture of the window. Therefore, the transmission factor of the film was always experimentally determined.

A thin polonium source was deposited on a silver button from a mother solution of RaD, E, F prepared by Dr. L. R. Hafstad, and was so arranged as to permit its accurate replacement with respect to the window holder. The number of alpha-particles passing through the aperture F in unit time without, and later with, the collodion film gave the ratio of the effective areas with and without the film. It is possible that for the lower energy of the fluorine alpha-particles (as compared with that of the polonium particles) the opacity was slightly different. However, its value obtained as just described always agreed closely with that estimated from the area of visual blackness seen under the microscope. This fact, together with the way in which the films were made, substantiates the belief that the opacity was due to the presence of "large" dust fragments and not to variations of film thickness. The correction was typically from two to four percent. The yields measured with different films were concordant. The same window frame and film were used at all angles in obtaining the angular distribution of the alpha-particles.

Solid-angle determination

The effective diameter of the small hole F was measured with a micrometer microscope to be 0.564 mm. Its mean distance to the center of the target-spot was determined with a micrometer depth gage to be 4.452 cm for the 90° port. These two measurements, together with a correction of one percent for the finite thickness of the small aperture and the finite size of the spot, resulted in a solid angle of $1.00 \pm 0.01 \times 10^{-5}$ of a sphere. It was slightly different at the other angles at which observations were made but in all cases was measured with similar accuracy.

This was the only geometric factor on which the results depended.

Targets

Thick natural crystals of calcium fluoride were used. Electrostatic charging of the crystal face[7] was reduced by a fine tungsten whisker which touched the face of the crystal just outside the bombarded area. Some charging was evident even then with a clean target surface but, in agreement with the experience of the Madison group, a few minutes' bombardment gave a sufficient layer of carbon to reduce the resistance drop along the surface to a negligible value.

Cleavage surfaces of the crystals were used. They were lightly scraped with a sharp knife at frequent time intervals to remove accumulated carbon. The surfaces were never ground or polished or brought into contact with any foreign matter which would be likely to become imbedded in them.

The possible effects of charging of the crystal and carbonization in reducing the effective bombarding voltage of the beam were guarded against by working at voltages sufficiently far

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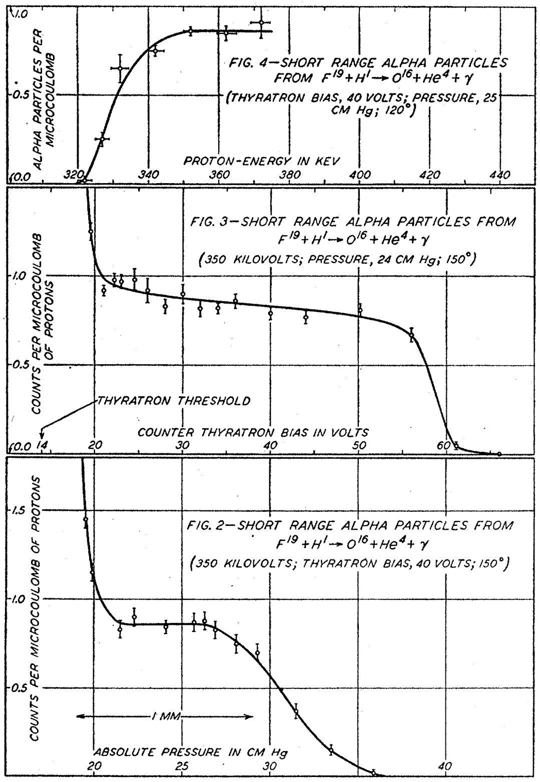
[7] E. J. Bernet, R. G. Herb and D. B. Parkinson, Phys. Rev. **54**, 398–408 (1938).



FIGS. 2, 3, and 4. The number of counts per microcoulomb of protons as a function of pressure in the absorption cell (Fig. 2), bias on the grid of the input tube of the scaling circuit (Fig. 3), bombarding voltage (Fig. 4). Exploratory curves of Figs. 2 and 3 served to determine optimum operating values of pressure and bias. In Fig. 2 the horizontal arrow marked "1 mm" indicates the change in pressure necessary to change the stopping power of the absorption region A by 1 mm of normal air at 15°C and 760 mm Hg. It can be seen that the region of complete separation of the alpha-particles and the scattered protons is about one-half of a mm air equivalent. The long range alpha-particles number about one-half percent of the short range alpha-particles and consequently do not appear here in measurable numbers.

above the 330-kv resonance to be safely on the horizontal portion of the yield-curve.

Two good quality CaF_2 crystals of different origin gave alpha-particle yields in agreement to within the accuracy of the measurements. A portion of one of them was examined for us by Dr. H. E. Merwin of the Geophysical Laboratory of the Carnegie Institution of Washington. He found that (1) the crystal was free from inclusions of foreign solid matter and of water and (2) its index of refraction differed less than 0.002 and its specific gravity less than 0.001 from other good specimens of mineral fluorite which he had selected over a period of several years. It can therefore be said that the crystals used in these measurements were representative of the best mineral crystals available. A quantitative chemical analysis for fluorine in fluorite is attended by such difficulties as to make any but a most painstaking analysis of very little value.

Targets prepared by the fusion or evaporation in air of compounds of fluorine were found to be unreliable for yield-measurements because heating the compounds in air reduced their fluorine content.

Angular distribution and yield measurements

The number of counts recorded by the amplifier-scaler-counter circuit per microcoulomb of bombarding protons was a function of three arbitrarily variable conditions:

(1) The pressure in the absorption cell.
(2) The bias on the grid of the input tube of the scaling circuit.
(3) The bombarding voltage.

The way in which the alpha-particle yield was experimentally found depended on obtaining plateaus in the curves of the counts per microcoulomb against the above three variables. These plateaus are shown in Figs. 2, 3, and 4. The curves are typical of a considerable number taken at 60°, 90°, 120° and 150° with respect to the forward direction of the proton beam.

From the heights of the horizontal positions of these three curves, taken together, was determined the number of alpha-particles per microcoulomb of incident protons. Such a value was obtained for each of the above-mentioned angles.

For each angle the crystal face was set so its normal approximately bisected the angle between the beam and the observation port. This minimized the effect of roughness of the target surface.

It was not easy to make measurements at smaller angles than 60° due to the obliquity of the crystal and the preponderance of scattered protons.

RESULTS

After having corrected the results at the several angles for the slight differences of geometry and the transmission-factor of the collodion film and for the motion of the center of mass, the values given in Table I were obtained. There is seen to be no systematic deviation from spherical symmetry. Thus the total yield over 4π steradians is

$$8.9\pm0.5\times10^4 \text{ short range alpha-particles}$$

per microcoulomb of 360-kv protons bombarding a thick crystal of CaF_2.

The probable error has been set equal to the square root of the sum of the squares of the errors introduced by the components described below. Our largest error we estimate to be that introduced in reading off the number of alpha-particles per microcoulomb from the data. In the number-thyratron bias curves the plateau obtained was never exactly horizontal and one has to use his best judgment in deciding what value to select. We took the uncertainty in this process to be three percent. The uncertainties in the current averaging, the collodion film transmission factor, the area of aperture F, and its mean-square distance from the target all combine to give a total probable error of five percent. The long range alpha-particles from reaction (*c*) were found to represent less than one-half percent of the total alpha-particle yield at 360 kv. No correction was made for them.

In view of the evidence cited above, the yield of 6.2-Mev quanta is taken to be equal to the alpha-particle yield.

Angular distribution of gamma-rays

In any experiment which does not use this radiation over the entire sphere it is important to know the angular distribution of the gamma-radiation. There are apparently sound theoretical reasons for expecting the radiation to be spherically symmetric. However, it was regarded as desirable to determine the actual distribution by measurement. The apparatus used for this purpose is shown in Fig. 5. The Geiger counter was placed in the heavy-walled lead box E. The box was mounted on a rigid framework whose axis of rotation passed through the target-spot and perpendicular to the beam. The front wall of E was sufficiently thick (6.4 cm) to reduce the intensity of 6.2-Mev radiation to about four percent of its incident value, except over the area of the aperture, which had the shape of a circular cone with vertex at the target. The geometry was such as to make $\Delta\theta=7°$. It was verified that this was the actual aperture by observing the effect of inserting a long brass plug in the conical hole.

TABLE I. *Alpha-particles per microcoulomb of bombarding protons observed at the various angles corrected for the motion of the center of mass and adjusted to the mean solid angle at 90°.*

ANGLE IN THE LABORATORY SYSTEM	COUNTS PER MICROCOULOMB IN 1.00×10^{-5} OF A SPHERE
60°	0.86 ± 0.10
90	0.87 ± 0.02
120	0.90 ± 0.02
150	0.89 ± 0.02

A Lauritsen electroscope was clamped in a fixed position with respect to the target and was used as a monitor for all measurements.

The angular distribution of the gamma-radiation from a thick target was determined for bombarding voltages of 370, 900 and 1000 kv. Reasons for selection of these voltages are evident from examination of the yield-curve.[7, 8]

At all three voltages the radiation was found to be spherically symmetric to within the experimental error of five percent.

Quantum intensity of gamma-radiation at 1050 Mev

As discussed above the gamma-ray energy is the same (with the exception of the slight amount of annihilation radiation described below) at all the resonances with which this work was concerned. Valid comparisons of the intensity of the radiation produced at various values of bombarding voltages can therefore easily be made with a Geiger counter or electroscope without attention to geometric conditions or to the nature or arrangement of the shielding. The ratio of the gamma-ray intensities at 370 kv and 1050 kv was carefully determined by simultaneous measurements with a Lauritsen electroscope and with a Geiger counter (Fig. 5). The two ratios always differed inappreciably.

Furthermore, the intensity ratio was found to be the same for moderate shielding (0.5 cm of lead over the sensitive area of the counter

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[8] L. R. Hafstad, N. P. Heydenburg and M. A. Tuve, Phys. Rev. **50**, 504–514 (1936).

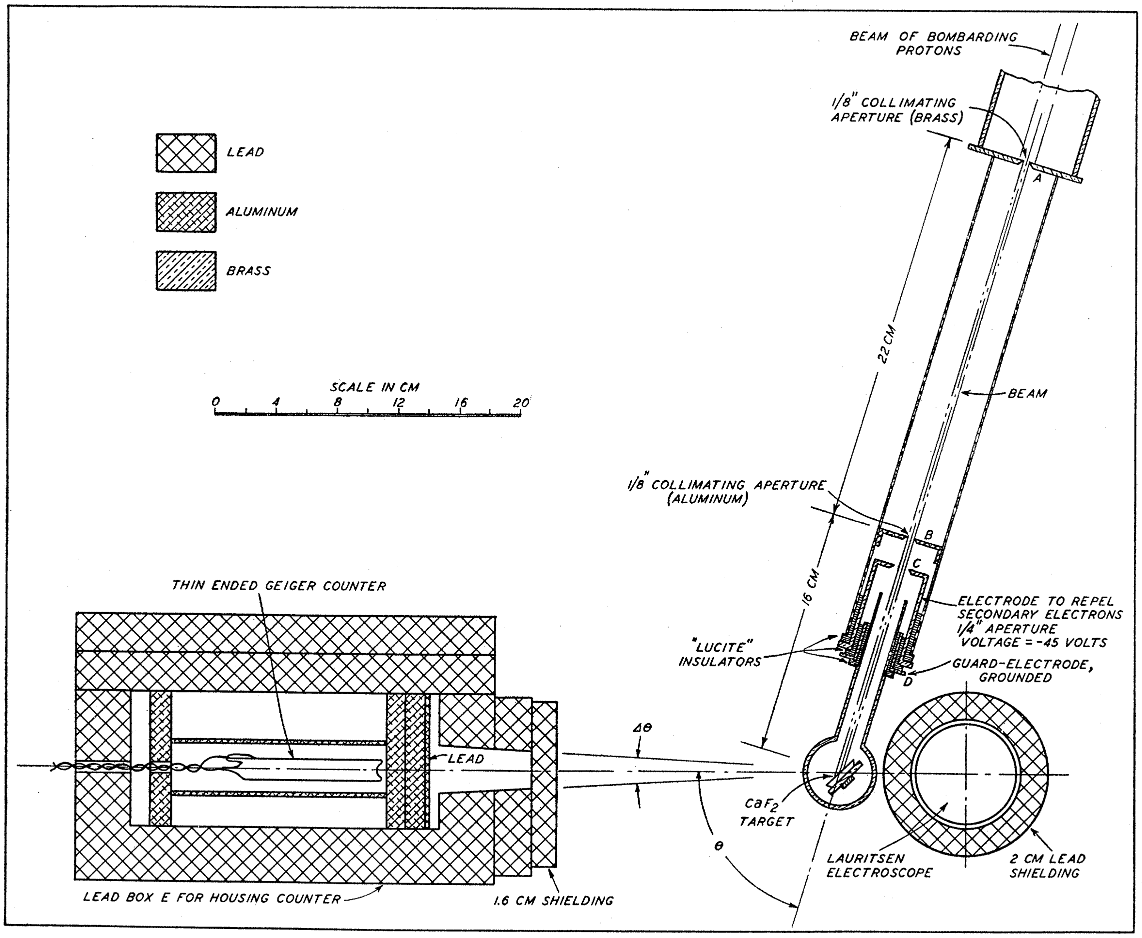

FIG. 5. Diagram of apparatus used to determine the angular distribution of gamma-radiation and the intensity at 1050 kv relative to that at 370 kv. For the angular distribution the lead marked "shielding" was not present. The ratio of the intensities was taken with and without the shielding with similar results.

and 0.3 cm of brass over the electroscope) and heavy shielding (2.0 cm lead for both the counter and electroscope). This assured that the results were not influenced by the presence of the slight amount of soft radiation from the annihilation of the positive members of the nuclear pairs which arise at the higher bombarding voltages.[9]

The average of all such determinations was

$$I_{1050\ \text{kv}}/I_{370\ \text{kv}}=42.0\pm0.8.$$

This is in good agreement with the earlier measurement (heavy shielding—one inch of lead) of 41.2 made in this Laboratory.[8]

Thus the yield of 6.2-Mev quanta from CaF_2 at 1050 kv is

$$3.7_4\pm0.2\times10^6 \text{ quanta}/\mu c \text{ of protons.}$$

DISCUSSION

A rough check on the absolute quantum intensity at 360 kv was made by comparison with a milligram of radium shielded with three cm of lead. The efficiency of the aluminum-sheathed Geiger counter for each line of the radium spectrum and for the fluorine radiation was taken to be proportional to the product of the true absorption coefficient of aluminum by

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[9] The Pasadena group estimates (private communication) the relative yield of pairs and gamma-ray quanta at 1 Mev to be approximately two percent. Since the sensitivity of detection of 6-Mev quanta is about four times as great as 0.5-Mev quanta, the maximum contribution of the annihilation radiation to the intensity ratio would be of the order of one percent for lightly shielded detectors and one-tenth percent for our heavily shielded ones.

the mean range of the secondaries.[10] Under these assumptions the number of quanta from the CaF_2 target per microcoulomb of protons is given by

$$Q_F = Q_{Ra}\left(\frac{C_F}{C_{Ra}}\right)\left(\frac{\sum_i p_i \tau_i R_i e^{-\mu_i x}}{\tau_F R_F e^{-\mu_F x'}}\right),$$

where C_F and C_{Ra} are the observed numbers of counts due, respectively, to one microcoulomb of protons bombarding fluorite and to one milligram-second of radium placed in the target position, p_i is the relative intensity of the ith gamma-ray line of the radium spectrum, the quantities $\tau_i R_i$ are proportional to the counter efficiencies mentioned above, the exponentials take account of absorption in the lead shielding, Q_{Ra} is the total number of quanta per second from a milligram of radium, and the summation is taken over the entire radium spectrum.

Using the best available values for all quantities involved in the above formula, and our ratio (C_F/C_{Ra}), we obtained a value for Q_F differing by less than 20 percent from the alpha-particle count. In view of the crudeness of this type of calibration the agreement is regarded as satisfactory.

It is to be noted that a similar calculation made by us on data previously published by Hafstad and Tuve[11] of this Laboratory gives 2×10^5 quanta/μc at 350 kv contrary—that is, about a factor of two too high instead of a factor of 1000 too small—to the commonly accepted interpretation of their data.[12]

The original estimate of McMillan[13] is in order of magnitude agreement with the intensity here reported.

Streib, Fowler, and Lauritsen[14] have recently made a Gray-Laurence type calibration of the fluorine source with an electroscope, whose sensitivity was determined by exposure to lightly shielded radium. Their thick-target result is 4.0×10^6 quanta/μc.

These measurements provide valuable independent checks of the quantum intensity, but in view of the complexity of the quantitative interpretation of ionization-chamber results, it appears that greater reliance can be placed on the direct alpha-particle count.

ACKNOWLEDGMENTS

We gratefully acknowledge the support of our Director, Dr. John A. Fleming, and the encouragement of Drs. M. A. Tuve, and L. R. Hafstad of this Laboratory. We are indebted to Dr. H. E. Merwin of the Geophysical Laboratory for the examination of one of the fluorite crystals used in this work. Professors C. C. Lauritsen and W. A. Fowler and Drs. J. F. Streib and R. A. Becker have discussed some of their experiments with us and have very kindly furnished in advance of publication recent data obtained in their laboratory.

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[10] See, for example, W. Bothe and W. Genther, Zeits. f. Physik **106**, 236–248 (1937).

[11] L. R. Hafstad and M. A. Tuve, Phys. Rev. **48**, 306–315 (1935).

[12] H. A. Bethe, Rev. Mod. Phys. **9**, 207 (1937).

[13] E. McMillan, Phys. Rev. **46**, 868–873 (1934).

[14] J. F. Streib, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **59**, 253–270 (1941).