

Proton-Gamma-Ray Coincidence Counting with Cyclotron Bombardment*

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A technique is given for studying associations between protons and gamma-rays emitted when a target is bombarded with particles from the cyclotron. A description is included of the theoretical considerations involved and of the apparatus developed. The results on the $Al^{27}(\alpha, p)Si^{30}$ reaction indicate that no gamma-rays accompany the end group of protons, while other groups exhibit coincidences. The coincidence rate increases with excitation energy. This may be due to cascade transitions giving more quanta, or to increased sensitivity for higher energy quanta. The fourth group observed with zero-degree bombardment is shown to be a composite of knock-on protons and protons from the reaction. A brief proton group study is included with Q values in essential agreement with previous work.

1. INTRODUCTION

ONE method of studying nuclear energy levels is the observation of proton groups from (α, p) and (d, p) reactions. These groups are usually ascribed to the formation of the residual nuclei in various energy states, the end group corresponding to the ground state. The nuclei which are excited then revert to the ground state with the emission of gamma-rays. If this interpretation and the energy levels derived from proton data are correct, coincidences should occur between gamma-rays and appropriate proton groups.

Because of the intrinsic difficulties involved, relatively little has been done on the gamma-radiation produced in such bombardment. Still fewer experiments have been carried out on coincidences between particles and gamma-rays emitted in transmutation reactions. Bothe and v. Baeyer¹ first observed proton-gamma-coincidences in the bombardment of boron by polonium alpha-particles. They found no genuine coincidences with the end group of protons, while the next group exhibited coincidences. Maier-Leibnitz,² using a paraffin "proton radiator" for the neutrons, found neutron-gamma-coincidences in the $Be^9(\alpha, n)C^{12}$ reaction. He and Bothe³ also

repeated the boron experiment. In these experiments natural alpha-particle sources were used.

The purpose of the present work has been to develop a technique by which proton-gamma-coincidence work could be done with cyclotron bombardment. The instrumentation problems for coincidence work with cyclotron bombardment are considerable, primarily because of the background radiation from the machine and because of the limitations on possible solid angles for the counters with respect to the target. The apparatus developed to meet these difficulties is described below with the first results obtained in the study of the $Al^{27}(\alpha, p)Si^{30}$ reaction. This reaction was chosen for developing the technique for two reasons. First, aluminum is a single stable isotope for which the proton groups from alpha-particle bombardment are well known. Second, the background radiation problem is smaller with alpha-particles than with deuterons. Once established, the method may be applied to other reactions.

The general procedure employed consisted of bombarding a thin target with particles from the cyclotron. Proportional counters, of appropriate designs, were used to record emitted protons and gamma-rays, respectively. Pulses from each counter were fed to a preamplifier and then to a high gain video amplifier. The amplified pulses were mixed in a coincidence circuit of special design, and coincidence pulses were recorded on a mechanical recorder.

Proportional counters were chosen for three reasons. First, since in the proportional counter

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¹ H. J. v. Baeyer, *Zeits. f. Physik* **95**, 417 (1935).

² H. Maier-Leibnitz, *Zeits. f. Physik* **101**, 478 (1936).

³ W. Bothe and H. Maier-Leibnitz, *Zeits. f. Physik* **107**, 513 (1937).

the pulses depend primarily upon the collection of electrons, the time lag uncertainty between the formation of the primary ionization and the occurrence of the counter pulse is smaller than with Geiger-Müller counters. This makes possible the use of fast electronic circuits without loss of genuine coincidences. Second, the proportional counter and its preamplifier recover very rapidly. Third, with a proportional counter for protons it is possible to set the counting level so that only those protons near the ends of their ranges will produce sufficient ionization to record in the counting circuits and Bragg "peaking" results. This allows one to examine each group separately in coincidence counting.

2. NUMERICAL CONSIDERATIONS

Most of the basic ideas involved in coincidence counting may be found in v. Baeyer's original paper.¹ Dunworth⁴ has treated the method in greater detail with special reference to radioactive decay schemes. Because of the occurrence of gamma-rays from the several competing reactions in target bombardment, and because peaking of the proton counter was employed, the equations which apply in the present case are somewhat different and more complicated.

Let N_p and N_γ be the counting rates per second in the two counters, C_o and C_e the genuine and chance coincidence rates per second, e_p and e_γ the intrinsic counter efficiencies, Ω_p and Ω_γ the counter-solid angles expressed as fractions of the

total sphere about the target, B the beam in particles per second, k_p , k_n , and k_s the yields per bombarding particle for the production of protons, neutrons, and inelastically scattered alpha-particles. Then

$$N_p = \rho_i k_p B e_p \Omega_p, \quad (1)$$

where ρ_i represents the fraction of the total number of protons produced that occur in the i th proton group. This factor appears because of the use of peaking. With this type of operation cosmic rays, gamma-rays, and beta-particles do not record in the proton counter. The background arising from neutrons in the proton counter is negligible when an alpha-particle beam is used.

Representing by W_p the average number of gamma-rays emitted per proton (protons of all energies are considered here), and by W_n and W_s the average number of gamma-rays emitted per neutron and per inelastically scattered alpha-particle, respectively:

$$N_\gamma = W_p k_p B \bar{e}_{\gamma p} \Omega_\gamma + W_n k_n B \bar{e}_{\gamma n} \Omega_\gamma + W_s k_s B \bar{e}_{\gamma s} \Omega_\gamma + \bar{e}_{\gamma b} F(B), \quad (2)$$

where $F(B)$ is the number of gamma-rays or cosmic rays incident upon the gamma-counter per second from sources other than the target. It is obviously some increasing function of B . Because the gamma-counter efficiency is a function of the gamma-ray energy, the e_γ 's in Eq. (2) must be interpreted as the weighted mean values of the efficiencies for *all* the different gamma-rays associated, respectively, with each type of particle. The additional subscript on the \bar{e} 's refers to the origin of the radiation, proton, neutron, etc. This expression obviously has too many unknowns to be of much value in itself. Its importance, however, lies in enabling one to write down the approximate Eq. (5).

If we assume that no *angular* correlation exists between associated protons and gamma-rays, the number of genuine proton-gamma-coincidences per second (corresponding to the i th proton group) is

$$\begin{aligned} C_o &= (\rho_i k_p B e_p \Omega_p) \Omega_\gamma \sum_{j=1}^n w_{ij} e_{\gamma j} \\ &= N_p \Omega_\gamma \sum_{j=1}^n w_{ij} e_{\gamma j}, \end{aligned} \quad (3)$$

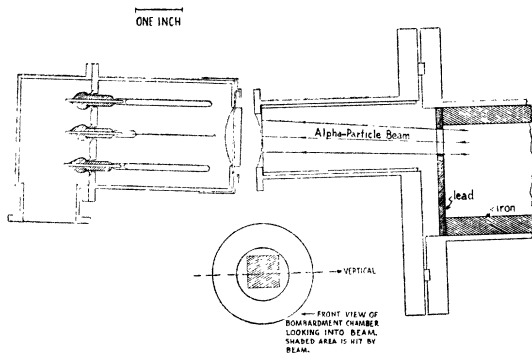
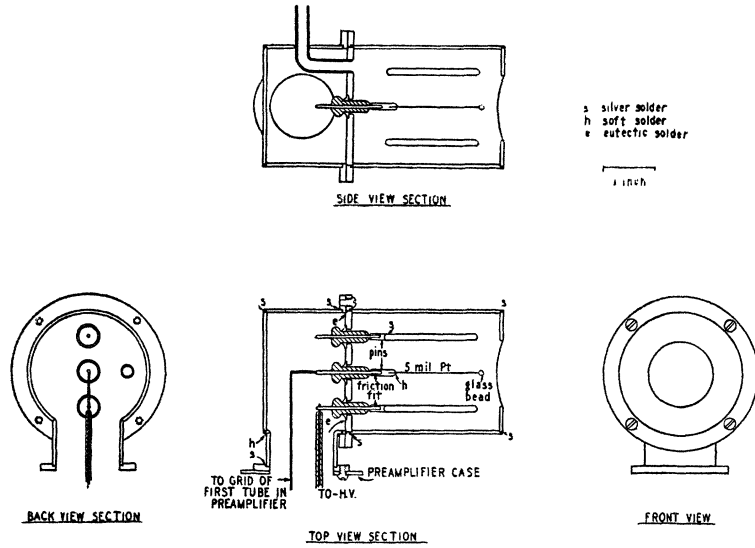


FIG. 1. Top view of the geometry for proton counting, showing, from right to left, the extension tube from the cyclotron, the rectangular limiting slit, the bombardment chamber and target, and the proton counter with its foil holder. The inset shows the target area hit by the beam.

⁴ J. V. Dunworth, Rev. Sci. Inst. 11, 167 (1940).

FIG. 2. Drawing of the details of the proton counter designed for large solid angle. The counter was connected directly to the preamplifier case.



where w_{ij} is the average number of gamma-rays of energy $h\nu_j$ associated with each proton in group i . Since "branching" may occur, w_{ij} may have any positive value, including fractional ones. The corresponding efficiency of the counting system for each gamma-ray is $e_{\gamma j}$. If branching does not occur, Eq. (3) becomes

$$C_o = N_p \Omega_\gamma w_i \bar{e}_\gamma, \quad (3a)$$

where w_i is the number of gamma-rays associated with each proton of the i th group, and \bar{e}_γ is the average efficiency for the gamma-rays associated with these protons. For the number of chance coincidences per second we have

$$C_c = 2TN_p N_\gamma, \quad (4)$$

where T is the resolving time of the counting circuit. Substituting the expressions for N_p and N_γ from (1) and (2) into (4), and dividing the resulting expression by (3a), we obtain

$$\frac{C_c}{C_o} \cong \frac{2TB}{w_i} [W_p k_p + W_n k_n + W_s k_s] + \frac{2TF(B)}{w_i \Omega_\gamma}. \quad (5)$$

The simplest case has been taken, with $\bar{e}_\gamma = \bar{e}_{\gamma p} = \bar{e}_{\gamma n} = \bar{e}_{\gamma s} = \bar{e}_{\gamma b}$.

Equation (5) and either (3) or (3a) are very useful qualitatively in designing the apparatus. In order to make C_o large, while keeping C_c/C_o small, it is desirable to have Ω_p , Ω_γ , e_p , and e_γ as large as possible. On the other hand, T should

be as small as possible, and the smallest beam intensity B consistent with yield is preferable. This minimizes not only the first term in (5) but also the second, since $F(B)$ is an increasing function of B . Good gamma-counter shielding is imperative to keep $F(B)$ small.

In a given experiment the total coincidence rate with the i th group of protons is measured, as well as N_p and N_γ . Since the proton counter is insensitive to cosmic rays, beta-particles, and gamma-rays, one needs only to calculate C_c from (4) and subtract this from the total coincidence rate to obtain C_o . Taking the ratio $C_o/(N_p \Omega_\gamma)$, one obtains a value for $\sum_j w_{ij} e_{\gamma j}$. This coupled

with a knowledge of the proton group energy spacing, and the gamma-counter efficiency *vs.* $h\nu$ curve, enables one to clarify the level scheme.

3. EXPERIMENTAL APPARATUS

The apparatus designed and constructed to fulfill as well as possible the above criteria is described below. A zero-degree bombardment chamber, Figs. 1 and 3, was chosen because of the large proton solid angle possible. The circular brass flange, into which was silver soldered the cylindrical brass tube, was bolted through a rubber-gasket vacuum seal to the tube leading to the cyclotron "can." Insulating bushings were used for the bolts so that the beam intensity could be measured by simply clipping a galvanometer lead to the chamber flange. The

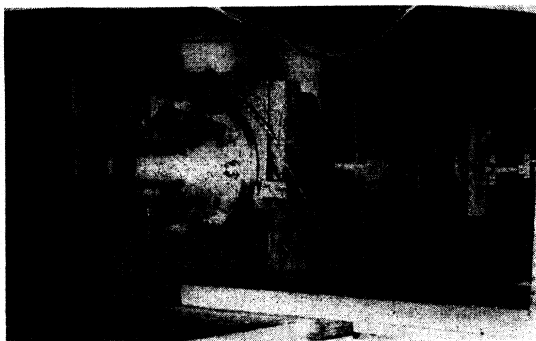


FIG. 3. Photograph showing the cyclotron magnet, with the extension tube leading out to the bombardment chamber. Part of the lead gamma-counter shielding is shown. The target was immediately behind the aluminum proton "window."

bombardment-chamber tube was made as small as possible to allow a large Ω_γ . An aluminum-foil (8.36-cm air—corrected for stretching under vacuum) proton window was waxed to the end of the chamber.

Immediately inside the window was placed a thin aluminum foil (1.01-cm air), backed by a gold foil of 8.60-cm air (stopping power taken as 5.20), which stopped the bombarding alpha-particles. These foils were carefully cleaned and were held in good metallic contact with the chamber by the inner friction tube shown in Fig. 1. The total bombardment chamber wall thickness through which gamma-rays had to pass was one-eighth inch. This will be reduced in future work to minimize absorption of weak

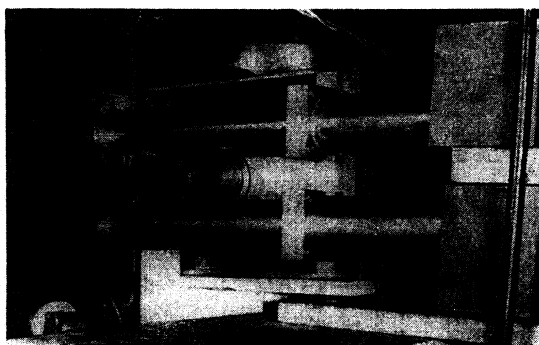


FIG. 4. Photograph of the gamma-counters in place above and below the bombardment chamber. A lead plate, for further shielding, was slipped over the nose of the bombardment chamber. The tube between each counter and its preamplifier was necessary to keep the latter out of the fringing field.

gamma-rays. The beam passed through a lead plate with a rectangular hole in it, which limited the beam to hitting the target only. The iron tube acted as a partial magnetic shield in getting the beam away from the machine.

The proton proportional counter, Figs. 1 and 2, was especially designed to give a large proton solid angle. The wire was 5-mil platinum with the unattached end melted into a tiny platinum ball and then coated with glass. This made the counter effective over nearly its entire cathode diameter. The large window was a 4.54-cm air (corrected for stretching) aluminum foil. The L-shaped shield, connecting the counter to the preamplifier case, was necessary because of space limitations near the cyclotron magnet yoke. The axis of the counter was parallel to the center of the beam.

Pure argon at 20 cm of Hg was used, with cathode potentials ranging from 740 to 1000 volts. The depth (beyond the window) to which protons had to penetrate in order to count (biases set for peaking) was calculated from data taken with ThC' alpha-particles to be approximately (0.6 ± 0.2) cm air. No attempt at great accuracy was made because of the poor proton-counting geometry necessary for the coincidence work. A foil holder was slipped over the nose of the counter so that carefully calibrated aluminum foils (1.518 mg/cm^2 equivalent to one cm air) could be placed between target and counter. The total basic absorption for the protons emitted at zero degrees was 25.1 cm air, subject, of course, to variation in stopping power with proton energy.

Because of the large collection angle for protons (the effective solid angle for coincidence counting was approximately 0.07) with the corresponding variation in both proton energies and basic absorption, extrapolated beam energies were used. Even then, how they should be extrapolated under the above conditions led to some uncertainty in measuring ranges. Clearly, this apparatus, designed for coincidence work, was not expected to give precision Q values. Those given below were obtained as a necessary by-product of the coincidence work.

The gamma-ray counter has been described in an earlier publication.⁵ A photograph of it with

⁵ B. B. Benson, *Rev. Sci. Inst.* **17**, 533 (1946).

expected here. A brief run was taken with the aluminum target removed, the beam hitting the gold backing. A proton yield was observed at an absorption corresponding to the fourth group and of the same order of yield. Another run was taken with a thin layer of vacuum grease smeared on the gold backing. A proton yield at the same absorption, but of approximately 100 times greater yield, was observed. The group consisted, therefore, at least largely of knock-on protons, although the coincidence work showed that some protons were truly associated with the aluminum reaction.

In Table I the extrapolated (and corrected) proton ranges, energies, and corresponding Q values from the present work are given, with the Si^{30} excitation levels derived therefrom. An extrapolated alpha-particle beam energy of 7.3 Mev was used. In addition, the Q values of Duncanson and Miller, Haxel, and Meerhaut are included.

II. In the coincidence work the voltage on the proton counter was set as high as possible (for high efficiency), subject to being somewhat below where beta-particles would record. The proton-yield curve with the counting system set as for coincidence counting is shown in Fig. 8. It should

be noted that the groups were separated, a fact which is important in the interpretation below.

The method employed in the coincidence studies was to set the proton absorption at each of the values indicated on Fig. 8 by the abscissae of the five vertical lines shown, i.e., 25-, 33.8-, 50.6-, 78.0-, and 89.7-cm air, respectively. The proton absorption was also set at "infinity" by inserting a $\frac{1}{16}$ -in. thick copper plate to stop all protons. This was a second control to check that the circuits were operating properly. At each of the absorptions the number of proton-gamma-coincidences per 1600 recorded protons was measured. The runs were taken in a random fashion so far as the value of the absorption was concerned, with an average of approximately 35-40 runs at each absorption. This method was chosen for three reasons: (1) Any slight fluctuations in counter efficiencies or cyclotron conditions canceled out. (2) The number of chance coincidences at each absorption was approximately the same, since although the proton counting rates at each absorption were different, those with low rates were counted on for longer times. (3) The number of genuine coincidences per proton allowed not only information on the gamma-counter efficiency to be found, but also

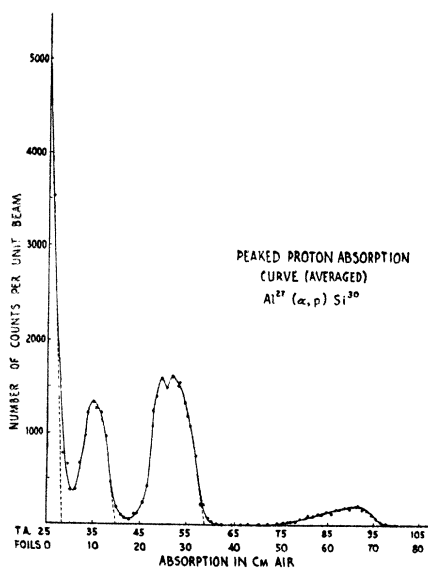


FIG. 7. Graph of the composite proton yield vs. absorption data, with biases set for the best possible peaking. The small dip in the second group occurred in each of the separate runs.

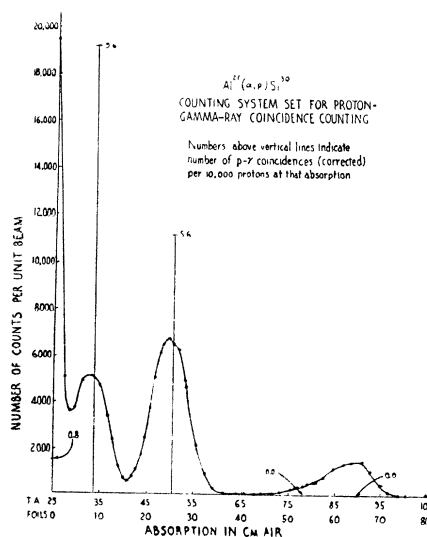


FIG. 8. Graph of the proton yield vs. absorption data, with biases set for coincidence counting. Compare the yields of Figs. 7 and 8. The numbers above the vertical lines indicate the number of genuine proton-gamma-coincidences per 10^4 protons at each of the five values of absorption.

TABLE II. Summary of the results of the proton-gamma-coincidence counting on the reaction $Al^{27}(\alpha, p)Si^{30}$.

Group	Absorption (cm air)	No. runs	No. protons	Ap-prox. total time (min)	No. coinc. C_{total}	Ex-pected chance coinc. C_c	Gen-uine coinc. C_g	Gen-uine coinc. per 10^4 proton	Probable error in $C_g/10^4$
1	89.7	32	51,200	64	3	3	0	0	—
1(a)	78.0	7	11,200	35	0	0.7	0	0	—
2	50.6	37	59,200	15	37	4	33	5.6	± 0.7
3	33.8	39	62,400	20	64	4	60	9.6	± 0.85
4	25.0	43	68,800	8	10	4.5	5.5	0.8	± 0.3
∞	∞	16	—	32	0	—	0	—	—

yielded information concerning gamma-ray transitions. The beam intensity was recorded each time (so that a knowledge of an average N_γ was possible) as well as the length of time for each run. All circuits were allowed to come to equilibrium before use. Table II is a summary of the results of the coincidence experiment on the aluminum reaction. The approximate probable errors were calculated by taking

$$\frac{(C_T)^{\frac{1}{2}}}{C_g} \times \frac{C_g}{10^4} \times 0.67.$$

5. DISCUSSION AND INTERPRETATION

It should be noted that in approximately 32 minutes of running (interspersed among the other runs), with an infinite absorption for the protons, no "coincidence" pulses occurred, indicating the circuits were operating satisfactorily. The number of expected chance coincidences was calculated in each case from Eq. (4), using a resolving time T of 5×10^{-7} sec. and an average N_γ equal to 65 per second (determined from the gamma-rate *vs.* beam calibration). Instead of considering N_γ as a counting rate, the total number of protons recorded was used, giving the number of chance coincidences directly.

The number of genuine gamma-ray coincidences observed with 51,200 protons in group one was zero, indicating that no gamma-rays are associated with the end group of protons. Because, however, of the much lower efficiency of a copper cathode gamma-counter for very low energy radiation,¹¹ one cannot exclude absolutely the possibility of a weak gamma-ray being associated with the end group. In addition to the much

lower efficiency any weak radiation would have suffered greater absorption in the wall of the bombardment chamber and in the counter case. From the coincidence work alone, therefore, the possibility of the association of a weak gamma-ray with the end group cannot be ruled out, but it can be stated that (except for the existence of a long-lived metastable state) the Si^{30} level corresponding to the end group cannot be more than a few tenths of a Mev above ground, for the occurrence of an energetic gamma-ray or several low energy ones in cascade would show up clearly. One means of clearing up this uncertainty would be to use a gamma-counter with a lead cathode, since its efficiency would not drop so greatly for low energies.

As further evidence, the high pressure ionization chamber work of Savel¹² in 1934 may be cited. He found that two components existed (one of 0.55 Mev and one of 2.1 Mev) in the gamma-radiation excited by the bombardment of aluminum by polonium alpha-particles. The former was attributed to annihilation and the latter to the (α, p) reaction.

Since the coincidence work excludes the possibility of any gamma-rays of greater than a few tenths of a Mev being associated with the end group, while other work indicates that no radiation of this order of energy is present (except for pair annihilation radiation), the conclusion to be drawn is that the end group leaves the Si^{30} in its ground state, and mass values calculated on this basis are justifiable.

The second group at 50.6-cm air gave 5.6 genuine coincidences per 10^4 protons. For lack of evidence concerning any excitation level in Si^{30} between 0 and 2.28 Mev, it is reasonable to assume that only one gamma-ray accompanies each proton. In this case Eq. (3) reduces to $C_g/N_p = \Omega_\gamma e_\gamma$, and putting in $C_g/N_p = 5.6 \times 10^{-4}$ and $\Omega_\gamma = 0.08$, we obtain $e_\gamma = 7 \times 10^{-3}$ for $h\nu = 2.28$ Mev. The intrinsic efficiency of the gamma-counter at 1600 volts is therefore approximately half that of a Geiger-Müller counter. With higher voltages the efficiency increases, being about 8.5×10^{-3} at 1650 volts.

For the third group (assuming no intermediate levels between 2.28 and 3.66 Mev) three possi-

¹¹ H. Bradt, P. C. Gugelot, O. Huber, H. Medicus, P. Preiswerk, and P. Scherrer, *Helv. Phys. Acta* 19, 77 (1946).

¹² P. Savel, *Comptes Rendus* 198, 368 (1934).

bilities exist: (1) A direct transition to ground with one gamma-ray of 3.66 Mev for each proton; (2) a cascade transition with one 1.38- and one 2.28-Mev gamma-ray accompanying each proton; (3) branching, with both types of transitions occurring. The expected number of genuine coincidences per 10^4 protons for the first two cases, assuming that no angular correlation exists and that the proportional gamma-counter followed the efficiency curve of a Geiger-Müller counter, can be calculated.¹¹ With $e_{2.28}$ equal to 7×10^{-3} , we get from simple proportions that $e_{1.38} = 4.1 \times 10^{-3}$ and $e_{3.66} = 9 \times 10^{-3}$. The latter is obtained by extrapolating the brass curve.

Therefore, in case 1, $C_g/N_p = 0.08 \times 9 \times 10^{-3} = 7.2$ coincidences per 10^4 protons. In case 2, $C_g/N_p = 0.08 \times (7 \times 10^{-3} + 4.1 \times 10^{-3}) = 8.9$ coincidences per 10^4 protons. The number of coincidences per 10^4 protons actually observed with the third group was 9.6 ± 0.85 . The calculated value from case 1 thus lies outside the probable range of fluctuation, while that for case 2 lies within it. This may be taken as indication that the cascade transition is more likely than the direct transition to ground. Since this interpretation is based upon the assumptions above, and depends, further, upon how the efficiency curve is extrapolated out to 3.66 Mev, the efficiency curve for the proportional gamma-counter must be determined experimentally before a conclusive statement can be made.

Finally, the value of 0.8 coincidence per 10^4 protons for the fourth group indicates that while most of the protons in this group are not associated with gamma-rays and are therefore knock-on protons, it is still possible to detect the presence of those protons associated with the (α, p) reaction. The brief data at 78.0-cm air are insufficient to make any quantitative statement, but the indication is that the first group is a single one.

6. SUMMARY

The first results of the application of the coincidence method to the study of the $Al^{27}(\alpha, p)Si^{30}$ reaction show that to a high degree of probability

the end group of protons leaves the Si^{30} in its ground state. The coincidences observed with the second group have been used to calculate the efficiency of the gamma-counter for 2.28-Mev radiation. Preliminary results indicate that a cascade gamma-ray transition from the second excited state in Si^{30} may be more probable than the direct one to ground. The presence of protons associated with the third excited state has been demonstrated, despite the high background of knock-on protons.

In considering further applications of the method to the study of nuclear transmutations, the following are possibilities: By producing the same residual nucleus in different ways, it is not only possible to see if the same levels occur in all cases, but also whether the same gamma-ray transitions occur. In this way information on any selection rules which may exist may be obtained. Further information may be obtained from the measurement of proton-gamma-coincidences per proton as a function of gamma-ray absorption, as well as from gamma-gamma-coincidence studies. The method should be applicable to the detection or examination of close nuclear "doublets" by measuring proton-gamma-coincidences per proton as a function of proton absorption. With variable delay techniques metastable levels may be looked for and studied. Reactions may be examined for angular correlations between protons and gamma-rays. The technique can be very useful in the interpretation of the proton groups obtained in the bombardment of multiple isotope elements.

7. ACKNOWLEDGMENTS

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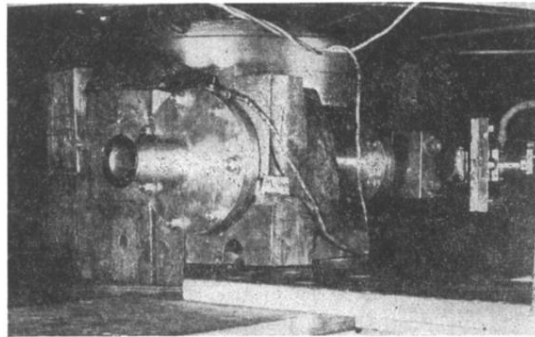


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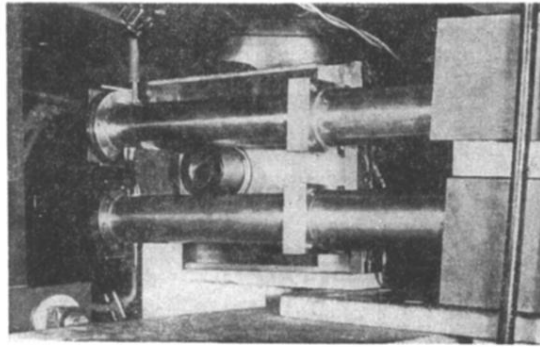


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