The Cross Section for the Radiative Capture of Protons by C^{13} at 129 kev*

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The cross section for the capture of protons by C^{13} at 129 kilovolts is

$$\sigma = 5 \pm 1 \times 10^{-33}\ \text{cm}^2.$$

This measurement was made possible through the use of a scintillation counter that had an over-all detection efficiency of 8.7 percent, and a pulsed ion source that had a peak proton current capability of one milliampere. A rough analysis of the radiation shows that 80 percent is due to the transition to the ground state of N^{14}, while the exact nature of the remaining 20 percent was not determined.

I. INTRODUCTION

THIRTEEN years ago H. A. Bethe proposed the carbon-nitrogen cycle as a source of solar energy, but the experimental evidence needed to verify or reject his theory was meager.[1] A program to obtain relevant evidence has been carried on for sometime in this laboratory. The excitation curves for the reactions in the cycle have been investigated over a wide range of proton energies, and the alpha-particle, gamma-ray, and positron spectra have been analyzed wherever possible. The cross sections become extremely small at solar energies ($\sim$25 kev) making it impossible to measure them directly. It has been considered desirable to make measurements at the lowest possible proton energies in order to reduce the range of the extrapolation required to estimate cross sections at such low energies.

All of the reactions, except the $C^{13}(p,\gamma)N^{14}$ reaction, give ionizing particles as reaction products. Such particles are more easily detected than gamma-radiation, and the cross sections for these reactions were the first to be measured at low energies.[2–4] The Geiger-Müller counter, ordinarily used in the past for detecting gamma-radiation, has two serious defects in low intensity measurements. First it is inefficient in converting gamma-radiation into detectable ionizing radiation since the amount of converting material is limited to the range of the secondary electrons produced. Second, because the pulse from the counter does not depend on the type of initiating radiation, x-rays and cosmic rays created a background that limits the intensity that can be observed. The scintillation counter partially overcomes both of these defects. The transparent crystal used as the phosphor has considerable mass, and will interact with a large part of the radiation passing through it. By placing the crystal close to the source a large solid angle is obtained without a complicated experimental arrangement. In addition, since the scintillation counter is a semiproportional detector, some of the background due to cosmic rays and x-rays can be eliminated.

The C^{12}, N^{14}, and N^{15} cross sections had been measured near 100 kilovolts, and it was considered desirable to measure the C^{13} cross section near this energy if at all possible. Estimates indicated that a proton current of one milliampere would be necessary to give a reaction intensity above the expected background. At the University of California a pulsed ion source has been recently developed that is capable of producing peak currents of this magnitude, but the average current is low, and if the advantage of a large peak current is to be utilized in overcoming background, the counter must be turned on only during the pulse.[5] This has been done without undue complication.

II. EXPERIMENTAL ARRANGEMENT

The accelerating voltage used in this experiment was supplied by a transformer and rectifier, while the accelerating column was a short version of the columns currently being used on the California Institute of Technology 3-Mev electrostatic accelerator. The ion source was a P.I.G. type ion source.[5,6] Figure 1 shows an assembly drawing of the source and focusing arrangement. Figure 2 shows a block diagram of the apparatus. The trigger source creates a 30 cps pulse which is lengthened to 760 microseconds by the counter-gate generator. The integrator anticoincidence circuit blocks this pulse, preventing operation of the ion source and scalers, during discharge of the ten microfarad integrating capacitor. After being passed by the anticoincidence circuit, the pulse starts the sweep of the current monitoring synchroscope, turns on the scalars by means of coincidence circuits, and finally is differentiated and converted into a light pulse. At the ion source this light pulse is reconverted into an electrical pulse that is used to start a single cycle multivibrator whose output pulse determines the duty cycle of the ion source. To provide proper integration the duty cycle of the scalers is made slightly greater than the duty cycle of the ion source.

* This work was assisted by the joint program of the ONR and AEC.

[1] H. A. Bethe, Phys. Rev. **53**, 608 (1938).
[2] R. N. Hall and W. A. Fowler, Phys. Rev. **77**, 197 (1950).
[3] Woodbury, Hall, and Fowler, Phys. Rev. **75**, 462(A) (1949).
[4] Schardt, Woodbury, and Fowler, Phys. Rev. **76**, 587(A) (1949).
[5] J. D. Gow, University of California, Berkeley, California (unpublished).
[6] A. Guthrie and R. Wakerling, *The Characteristics of Electrical Discharges in Magnetic Fields* (McGraw-Hill Book Company, Inc., New York, 1949), Chapter on "The P.I.G. Type Discharge."

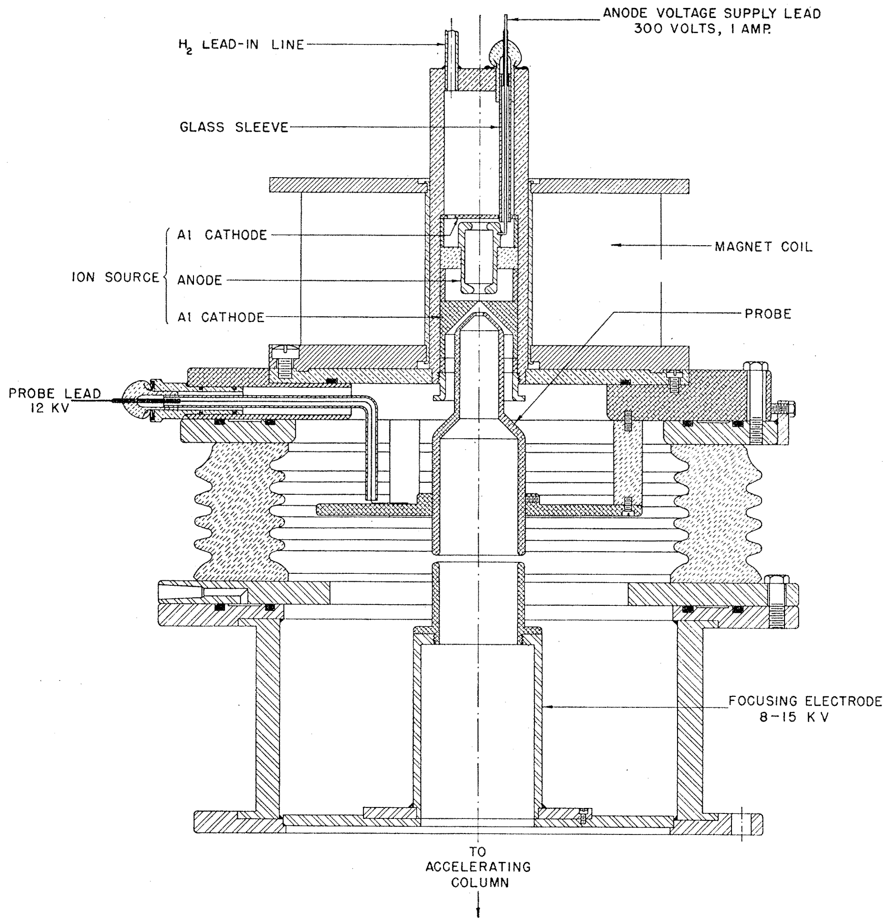

FIG. 1. Philips Ion Gauge (P.I.G.) type discharge ion source, and focusing system used with the accelerator.

The accelerating voltage was determined by measuring the current in a precision resistor column with a Weston microammeter. The actual voltage available for accelerating the protons is lower than the meter reading, for it depends on the current drawn from the voltage supply during the pulse, whereas the meter reading depends on the average current. Because the pulse current amounts to two milliamperes in addition to the average current of one and one-half milliamperes, and because the internal resistance of the supply was purposely made 500,000 ohms in order to limit the current during the occasional breakdowns of the accelerating column, it is necessary to make a one kilovolt correction to the meter reading. In addition to the 0.25 percent

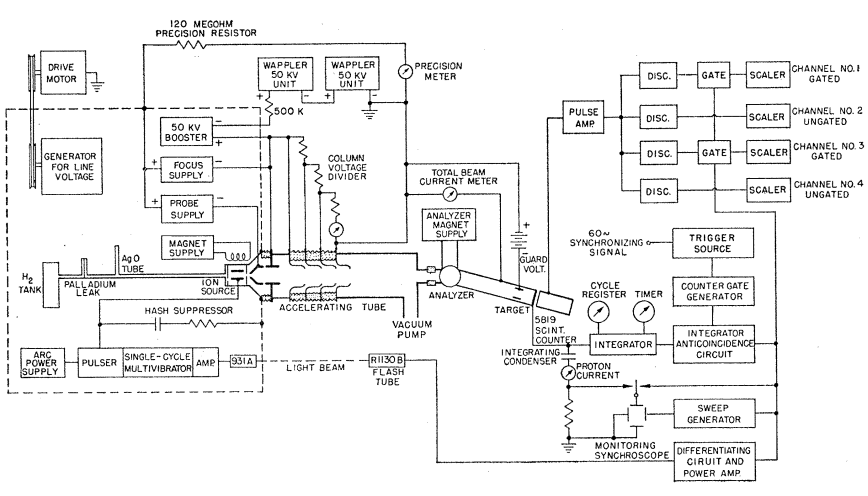

FIG. 2. Block diagram of apparatus used in the 128 kilovolt determination.

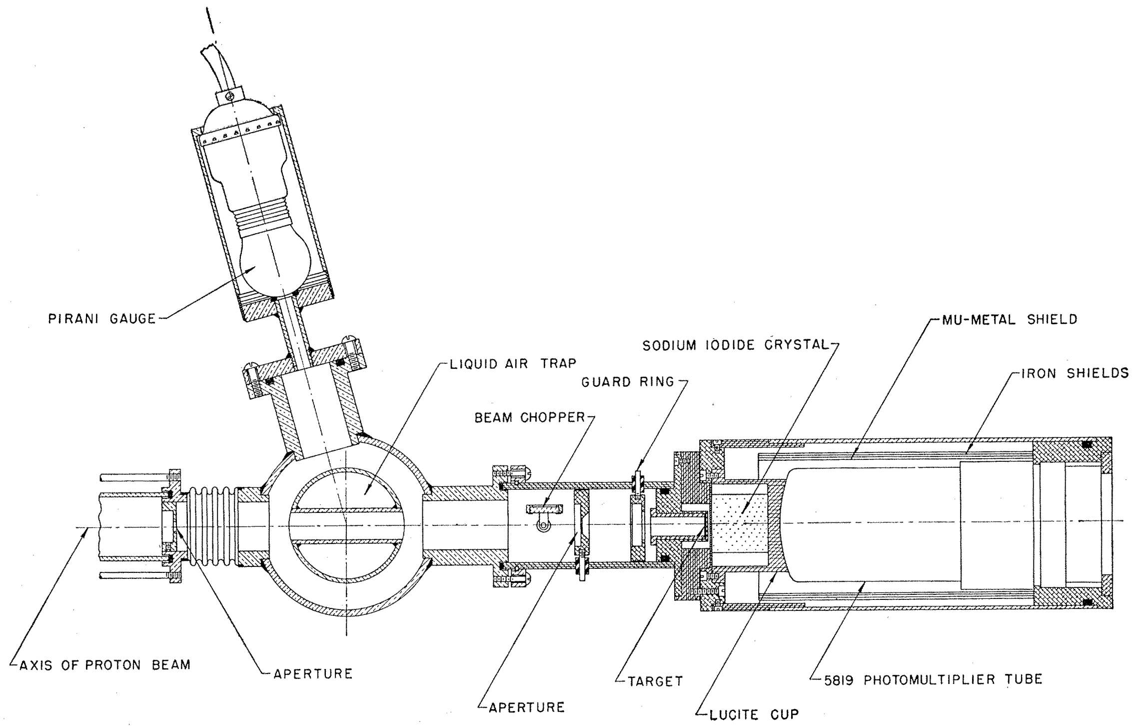

FIG. 3. Scintillation counter and target chamber. The target chamber vacuum was ordinarily 2×10^{-5} mm or better. The Pirani gauge is merely for rough vacuum readings when pumping down after target changes.

error in the voltage measurement an uncertainty of ~1 kilovolt is introduced by the ripple and fluctuations of the accelerating voltage.

III. COUNTER CONSTRUCTION AND CALIBRATION

Figure 3 shows an assembly drawing of the counter and target chamber. The distance between the target surface and the face of the thallium-activated sodium iodide crystal is 0.040 inch. The crystal is a one-inch cube cut from a larger blank supplied by the Harshaw Chemical Company. Because of the hygroscopic nature of sodium iodide, the Lucite cup is filled with sodium-dried mineral oil. A layer of Celvacene grease is formed between the Lucite cup and the photosensitive cathode of the 5819 photomultiplier tube to improve the optical coupling.

Photomultipliers and electronic amplifiers are subject to a certain amount of inherent instability. Figure 4 shows the integral bias curve obtained from a Co^{60} source that could be placed in a standard position near the counter. By finding the intercept of the straight line portion with the ordinate a convenient check of the detection efficiency is made, while the intercept of the straight line with the abscissa is a measure of the amplification of the system. The break at low bias is due to back-scattered radiation from Compton interactions in the surroundings of the crystals. With the amplifier gain reduced, the integral bias curves shown in Fig. 5 were found for gamma-rays of four different energies. Figure 6, showing the relation between the x-intercepts of the straight line portions of these curves and the energy of the respective gamma-rays, demonstrates the semiproportional nature of the scintillation detector. It is possible to obtain gamma-ray energies more accurately through the use of a differential technique, but such a technique is not an efficient way to make weak intensity measurements. Neglecting the radiation scattered into the crystal from the surroundings, the y-intercept of an integral bias curve gives the fraction, that is counted, of the yield of the source into the solid angle subtended by the counter. It can be shown, under reasonable assumptions, and if the points corresponding to less than a few hundred kilovolts of energy are neglected, that the error made by neglecting the scattered radiation is fairly small for the radiation energies considered in this experiment. Furthermore, any such error is partially eliminated if the absolute calibration of the counter is made using a gamma-ray energy comparable to gamma-ray energies in the unknown source.

The absolute calibration of the counter was made using the radiation from the $F^{19}(p,\alpha\gamma)$ reaction at one million volts bombarding energy. The radiation from this reaction consists of a mixture of 6- and 7-Mev gamma-rays, and at this bombarding energy the thick target yield for a thick CaF_2 target in the forward direction is 7.1×10^{-7} gamma-rays per proton.[7] There are also 5-Mev nuclear pairs from this reaction, and the

[7] Chao, Tollestrup, Fowler, and Lauritsen, Phys. Rev. **79**, 108 (1950).

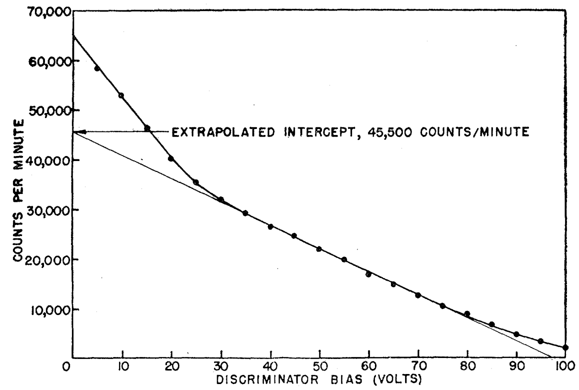

FIG. 4. Integral bias curve obtained using the calibrating Co^{60} source. The amplifier gain is eight times that of Fig. 5.

absorber between the target and crystal was not sufficient to absorb them entirely. The yield of the nuclear pairs is given as 0.10×10^{-7} particle per proton.[8] Therefore, taking into proper account the increased counter efficiency for particles over gamma-rays, the effective yield of the reaction for this counter is determining its detection efficiency for gamma-radiation is 7.4×10^{-7} event per proton. The experimental yield was found to be 422,000 counts for 1.05 microcoulombs of protons. Over a limited range of gamma-ray energies the efficiency of the counter might be expected to be proportional to the total absorption coefficient of sodium iodide. In an experiment to check the validity of this assumption the yield of the 2.3-Mev radiation from $C^{12}(p,\gamma)$ was determined at one million volts bombarding energy using a graphite target. It was found to be 7.2×10^{-10} gamma-ray per proton as compared to the value 7.3×10^{-10} given by other methods.[9,10] This required a 12 percent correction because of the change in the total absorption coefficient in going to the lower energies, and so the close agreement between the yield found and that predicted, justifies the assumption made.

The C^{13} targets used in the low energy work were prepared by cracking enriched methyl iodide on clean tantalum strips. The resulting layer of carbon was thin and hard.[11] The actual percentage of carbon as C^{13}, 61 percent according to the data supplied by Eastman Kodak, was checked by comparative yield measurements with ordinary graphite. Measurements made at the sharp 1.76-Mev resonance in C^{13} indicated that the thickness of the targets for protons was 20 kev at this energy. Because of the increase in the stopping cross section for carbon at lower energies, the thickness for protons at the energy used was great enough to insure a thick target yield.

[8] Streib, Fowler, and Lauritsen, Phys. Rev. **59**, 253 (1941).
[9] Fowler, Lauritsen, and Lauritsen, Revs. Modern Phys. **20**, 236 (1948).
[10] Recently a value of 7.7×10^{-10} has been found by J. D. Seagrave of this laboratory for this yield.
[11] J. D. Seagrave (to be published).

IV. REDUCTION OF THE DATA FROM $C^{13}+H^1$

After shielding the counter with two inches of lead, a good fraction of the background pulses, presumably those due to high energy cosmic-ray particles, were larger than any that could arise from the C^{13} reaction. This information was used to reduce the uncertainty in the background correction. Also it is possible and desirable to utilize the period between proton pulses for measuring the background, for then local fluctuations are more likely to be accounted for. With these ideas in mind the data were taken with four scalers and four discriminators. The first scaler was turned on only during the proton pulse and its discriminator was set to the bias voltage at which a reading was desired. The second scaler was on all the time and its discriminator was set to the same bias voltage as the first. The third scaler was turned on only during the proton pulse and its discriminator was set to a voltage just larger than the largest pulse that could arise from the reaction. The fourth scaler was on all the time and its discriminator was set to the same bias the third. We designate the counts collected by these scalers as C_1, C_2, C_3, and C_4, respectively. The data C_3 and C_4 may be subtracted from C_1 and C_2 for they represent pulses which are known to be too large. The numbers (C_1-C_3) and (C_2-C_4) then form the raw data to be used. We designate these by n_0 and N_0, respectively. The quantity (N_0-n_0) contains only the background for a period proportional to $(1-\tau)$, where τ is the duty cycle. Therefore, B, the background while the counter is on, is

$$B=\tau(N_0-n_0)(1-\tau)^{-1}.$$

The counts, n, corresponding to the C^{13} radiation are therefore given by

$$n=n_0-B=(n_0-\tau N_0)(1-\tau)^{-1}.$$

The statistical error in n is computed by noting that the error in $(N_0-n_0)(1-\tau)^{-1}$ is $0.7(N_0-n_0)^{\frac{1}{2}}(1-\tau)^{-\frac{1}{2}}$, and

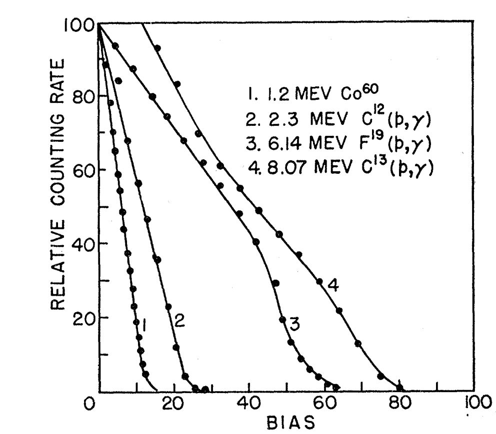

FIG. 5. Integral bias curves with targets of calcium fluoride, graphite, and enriched C^{13}. The break in the C^{13} curve is due to a lower energy component in the radiation. The Co^{60} curve of Fig. 4 is shown replotted to the same scale as that of this figure.

so the error in B can be found by using the differentiation rule and simplifying the resulting expression to yield

$$\epsilon(B)=0.7\tau(1-\tau)^{-1}(N_0-n_0)^{\frac{1}{2}}(1+\tau)^{\frac{1}{2}};$$

therefore the statistical error in n is

$$\epsilon(n)=0.7[n_0+\tau(1-\tau)^{-1}(1+\tau)(N_0-n_0)]^{\frac{1}{2}}.$$

If $\tau\ll 1$, and if $n_0<N_0$, as is true in this experiment then the error may be written as

$$\epsilon(n)\approx 0.7(n_0+\tau N_0)^{\frac{1}{2}}.$$

The yield can be determined by using the counter calibration obtained from fluroine, or it can be determined, assuming the yield as known at 700 kev, by comparing the counter yield at the lower voltage with the counter yield at 700 kev. When the yield has been calculated the cross section can be found by using the method of Hall and Fowler.[2] This yields the equation,

$$\sigma=(3Y\epsilon/fE^{\frac{3}{2}})(1+E^{\frac{1}{2}}/Z_0),$$

where f represents the fraction of C^{13} in the target, ϵ the stopping power of carbon, and E the energy of the bombarding protons in Mev. This equation is found by evaluating the following integral assuming that ϵ is constant,

$$Y=\int_0^E \frac{\sigma}{\epsilon}dE.$$

The assumption is good, for ϵ is nearly constant from 90 kilovolts to 130 kilovolts, and σ drops so rapidly with energy that the contribution below 90 kilovolts is relatively unimportant. Hall and Fowler give as an approximate form for σ,

$$\sigma=(a/E)\exp(-0.99Z_0/E^{\frac{1}{2}}).$$

This is derived from the asymptotic form of the penetration factor.

Typical counting data are tabulated in Table I and

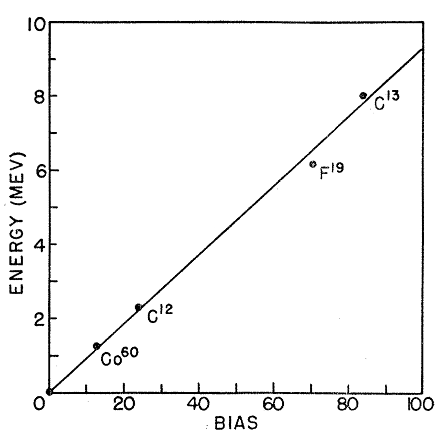

FIG. 6. The bias energy relationship for the data of Fig. 5 using the extrapolated intercepts of the straight line portions with the abscissa as the bias values.

TABLE I. Data obtained from C^{13} at 129 kilovolts.

Bias	C_1	C_2	C_3	C_4	Charge microcoulombs	True counts per 1000 microcoulombs
10	63	5	643	182	26,400	1.84±0.22
15	66	6	484	230	35,200	1.54±0.16
20	65	4	602	303	48,400	1.21±0.13
25	61	5	512	282	46,600	1.09±0.12
30	73	13	745	454	70,400	0.759±.081
40	44	9	456	326	48,400	0.665±0.089
50	21	3	314	228	38,700	0.417±0.084

Voltage 129±1 kilovolts
$\tau=0.0227$

shown graphically in Fig. 7. Making use of the counter efficiency of 8.7 percent, the target enrichment of 61 percent, and the y intercept from Fig. 7 corresponding to the hard radiation, the yield of the hard radiation for a pure target is found to be, 3.7×10^{-15} gamma-ray per proton. Noting that the soft radiation is about 3.5 Mev the yield of soft radiation is 2.8×10^{-15} gamma-ray per proton for a pure target. The corresponding cross sections are

$$\sigma=3.7\times 10^{-33}\text{ cm}^2$$

and

$$\sigma=2.8\times 10^{-33}\text{ cm}^2$$

respectively. Assuming the soft radiation is due to a two-step cascade a total cross section may be written

$$\sigma=5.1\times 10^{-33}\text{ cm}^2$$

or assuming a three-step cascade,

$$\sigma=4.6\times 10^{-33}\text{ cm}^2.$$

In either case we can round off the results to give the cross section as $5\pm 1\times 10^{-33}$ cm² at 129±1 kev.

Figure 8 gives a bias curve made using an ordinary graphite target at a bombarding energy of 700 kilovolts. The soft radiation is mostly from C^{12}. Comparing the yield of the hard radiation from this target at 700 kilovolts with the yield of hard radiation from the enriched target at 129 kilovolts, gives

$$Y_{700}/Y_{129}=2.2\pm 0.3\times 20^6.$$

The errors have been estimated as follows. In the ratio just given only the counting rates, the values of the charges collected by the integrator, and the enrichment factor enter directly. The integration error is negligible. The statsitical and extrapolation error in the counting rate at 700 kilovolts is not more than a few percent, and at 129 kilovolts it is ten percent. The enrichment factor is known within a few percent. Therefore the probable error has been set at 12 percent.

In determining the absolute cross section other errors arise. The fluorine yield which was used for calibration is uncertain to ten percent, and the stopping cross section which enters into the calculation is not known to better than ten percent. These errors, combined with

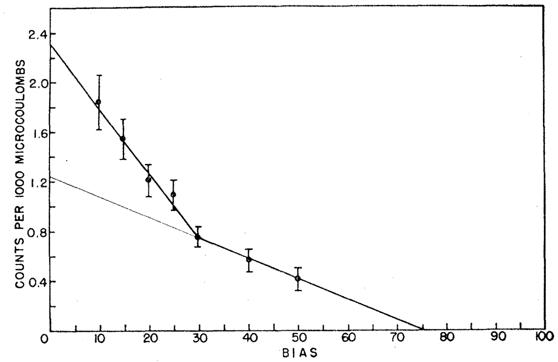

FIG. 7. Integral bias curve at 129 kilovolts for C^{13} using data of Table I.

those already mentioned, produce an uncertainty of twenty percent in the cross section.

The error in the accelerating voltage has been given as ~1 kilovolt. If surface layers were formed on the target, the effective accelerating voltage might be much lower than calculated. Because of the very rapid variation of cross section with energy, a one percent reduction in the voltage would result in an eight percent reduction in the yield. If a surface layer built up on the target, the counting rate would rapidly decrease in each experiment. This was not observed. In addition the targets did not show any discoloration after use.

V. COMPARISON WITH THE RADIATION AT HIGHER ENERGY

It is possible to make some prediction of the cross section to be expected at 129 kilovolts from the nature and yield of the reaction at higher energy. There are many resonances, but only two are of sufficient strength and breadth to contribute to the cross section at 129 kilovolts. These are the well-known resonance at 554 kilovolts, and a broad resonance at 1.25 million volts.

It is not known whether these resonances will interfere at the low energy, and so for comparison with the observed results they will be studied separately. The most useful tool for doing this is the single level Breit-Wigner dispersion formula,

$$\sigma=\pi\lambda\!\!\!^{-2}\frac{\omega\Gamma_\gamma\Gamma_p}{(E-E_R)^2+\frac{1}{4}\Gamma^2}.$$

$\lambda\!\!\!^{-}$ is the wavelength of the incident proton in the center-of-mass system, $\omega\Gamma_\gamma$ is a small factor nearly independent of energy,[12] E_R is the energy of the bombarding proton at resonance, E is the actual bombarding energy, Γ is $\Gamma_\gamma+\Gamma_p$, and finally Γ_p is given by $P(E)^{\frac{1}{2}}G$ where $P(E)^{\frac{1}{2}}$ is the penetration factor while G is the width at one million volts without barrier. $P(E)^{\frac{1}{2}}$ has been tabulated by Christy and Latter as a function of target nucleus and bombarding energy.[13] At low energies Γ_p becomes very small and thus Γ may be neglected in comparison to $(E-E_R)$. For purposes of extrapolation the equation

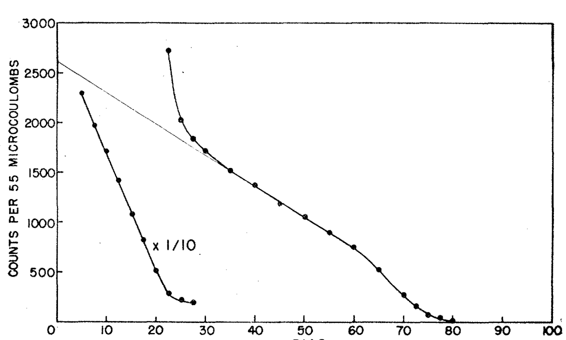

FIG. 8. Integral bias curve at 700 kilovolts for a graphite target.

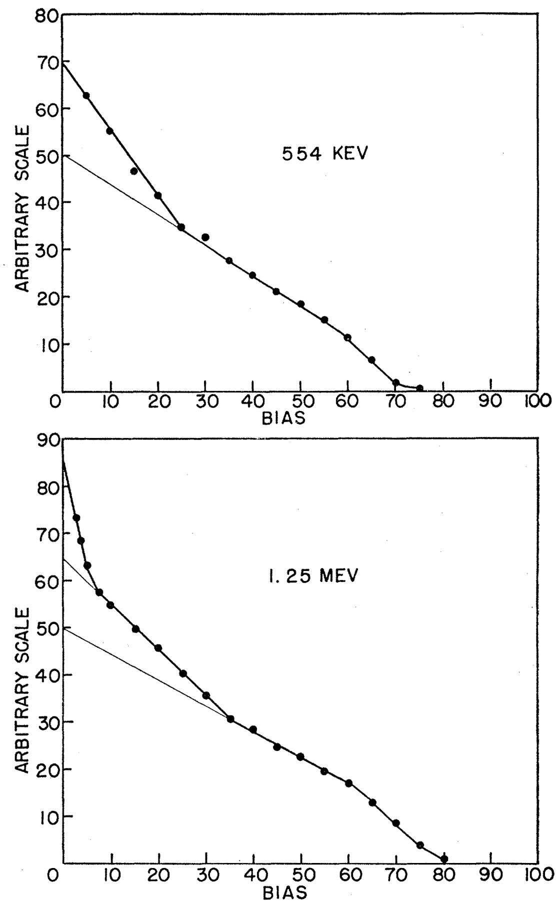

FIG. 9. Integral bias curves at 554 kilovolts and 1.25 million volts using thin enriched C^{13} targets.

[12] Strictly speaking Γ_γ is proportional to E_γ^3 for dipole radiation. For high energy gamma-rays and comparatively small variations in bombarding energy the statement made in the text is true. The error made by neglecting the dependence of Γ_γ on energy is not greater than 10 percent for the worst case.

[13] R. F. Christy and R. Latter, Revs. Modern Phys. **20**, 185 (1948).

may be re-expressed as

$$\sigma=\frac{E_R}{E}\frac{P(E)^{\frac{1}{2}}}{[P(E)^{\frac{1}{2}}]_R}\left[\frac{\Gamma_{pR}}{E-E_R}\right]^2\frac{\sigma_R}{4},$$

where $\pi\lambda\!\!\!^{-2}$ has been given by

$$\pi\hbar^2M/2\mu^2E.$$

Here μ is the reduced mass of the system, and M is the proton mass.

The integral bias curves at 554 kev and also at 1.25 Mev taken with a thin target are given in Fig. 9. Except for the very low energy tail on the higher energy curve, they are of the same nature as the curve at 129 kev. They both show about 35 percent total soft radiation as compared to 43 percent for 129 kev, but the error on the latter figure is sufficient to rule out attaching any importance to the difference. J. D. Seagrave has found the widths of the resonances as 32.5 kev for the lower resonance and 500 kev for the higher resonance. The best values available for σ_R are $\sigma_{554}=1.44\times10^{-27}$ cm² and $\sigma_{1.25}=0.062\times10^{-27}$ cm². These are calculated from total yields using Geiger tubes.[11] Making use of the extrapolation formula these yield

$$\sigma=2.55\times10^{-33}\ \text{cm}^2$$

and

$$\sigma=0.99\times10^{-33}\ \text{cm}^2,$$

respectively, at 129 kilovolts. This assumes both resonances are due to s-wave protons. These may be combined to give

$$\sigma=3.54\times10^{-33}\ \text{cm}^2$$

assuming no interference. It is seen that this value as well as the bias curves would emphasize the cascade as predominantly a triple one, but the evidence is not strong.

R. G. Thomas has recently proposed an improved method of extrapolation in place of the method used here.[14] His results are difficult to apply except in the case of C^{12} and similar nuclei with spin zero in the ground state, but they indicate that the cross section predicted by the standard method should be too low. In the case of C^{12}, which has been measured by Hall and Fowler, his method indeed predicts the correct value. In this regard it is interesting to note that the ratio of the cross sections for the capture of protons by C^{13} to C^{12} at 129 kilovolts is 4.6, and is the same as that predicted by the extrapolation from higher resonances despite the fact that both cross sections are thirty percent higher than the extrapolated values.

We wish to acknowledge the assistance of J. D. Seagrave and R. B. Day in operating the 3-Mev electrostatic generator for the fluorine calibration and other work done at higher energies. In addition we should like to thank the entire staff of the laboratory for the many suggestions and services that they rendered.

[14] R. G. Thomas, Phys. Rev. **81**, 418(L) (1951).