THE RICE INSTITUTE

THE (n,2n) CROSS-SECTION AT 14.1 MEV FOR
\( ^{14}N, \; ^{23}Cl \), AND \( ^{19}F \)

BY

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INTRODUCTION

This experiment is concerned with a method of determining cross-sections of \( (n,2n) \) reactions, taking advantage of the fact that the residual nucleus in most cases is a positron emitter which permits detection by coincidence counting. In this paper, the \( ^{14}\text{H}(n,2n)^{13}\text{H} \), \( ^{35}\text{Cl}(n,2n)^{34}\text{Cl} \), and \( ^{19}\text{F}(n,2n)^{18}\text{F} \) reactions are considered. These reactions have \( Q \)-values of -10.9, -13.2, and -10.3 mev respectively, and may be studied with the 14.1 mev neutrons which are produced by the \( ^{2}(d,n)\text{He}^4 \) reaction by the Rice Institute Cockcroft-Walton Accelerator. The \( ^{13}\text{N} \), \( ^{34}\text{Cl} \), and \( ^{18}\text{F} \) nuclei decay by positron emission with half-lives of 10.2 min, 33 min, and 112 min respectively.

The procedure of the experiment involves irradiating the substance under consideration for a measured time with a known flux of 14.1 mev neutrons, and then observing the coincidences due to the annihilation photons with an appropriate detecting system. The difficulties involved in calculating solid angles, efficiencies of the counters, etc., are avoided by calibrating the system in terms of a \( \text{Na}^{22} \) solution, which is absolutely calibrated previous to the experiment. The number of coincidence counts produced by the radioactive substance over a measured period of time is then easily related to the cross-section for the reaction.
DETERMINATION OF THE CROSS-SECTIONS

The experiment was first planned with the view of investigating the $\text{H}^{14}$ reaction, for which the cross-section has been reported to be 0.265 millibarns by Cohen\textsuperscript{2} for neutrons in the range 10-18 mev and 5.67 millibarns by Paul and Clarke\textsuperscript{3} for neutrons of 14.5 mev. In view of the small values for the cross-section, it was necessary to choose a favorable geometry for irradiation and counting. Liquid nitrogen contained in a spherical Dewar flask surrounding the neutron source appeared to be an effective arrangement for providing a large yield of $\text{H}^{13}$ nuclei. However, the yield per unit volume of the sphere falls off inversely as the square of the radius, so that the liquid on the outer layers of the sphere dilutes the activity of the more intensely irradiated material close to the neutron source. Therefore, it was necessary to choose a volume large enough to provide a reasonable total yield of radioactive nuclei, and yet small enough to provide high concentration per unit volume, so that coincidence counting would be efficient with the available detectors. Since a spherical Dewar flask of suitable dimension was not available, a hemispherical lucite vessel was made with a volume approximately satisfying the above requirements. This container was kept submerged in a cylindrical Dewar flask filled with liquid air for cooling and insulation. (See Figure 1) The apparatus was found to be impractical for the subsequent coincidence counting, because of excessive absorption of the annihilation photons in the source. To reduce this effect, the nitrogen was irradiated in the lucite container as described, and then poured
FIGURE I - THE IRRADIATION APPARATUS
into a previously cooled Dewar flask which was made in the form of a test-tube. This apparatus was also suitable for use with other materials available in liquid form and was used in this experiment with carbon tetrachloride for the $\text{Cl}^{35}$ reaction, and with Kel-F polymer oil for the $\text{F}^{19}$ reaction.

A. Derivation of the Equation for the Cross-section.

A hemispherical volume of the substance under consideration is irradiated, with the neutron source at the center of the hemisphere. The yield of radioactive nuclei in a hemispherical shell of thickness $dr$ at radius $r$ will then be

$$dY = \frac{J \rho A}{2M} dr$$

where $J$ is the total number of neutrons produced from the target per second, $\rho$ is the number of nuclei per cm$^2$ in the shell, and $\sigma$ is the cross-section for the reaction. In the case of liquid nitrogen is used to provide a large concentration of nuclei, $\rho$ is given by $\int_0^R \rho A d r$, so that

$$dY = \frac{J \rho A \sigma R}{2M} dr$$

where $\rho$ is the density of the nitrogen, $A$ is Avogadro's number, and $M$ is the atomic weight of the $\text{N}^{14}$. Then, by integrating over the entire hemisphere,

$$Y = \frac{J \rho A \sigma R}{2M}$$

where $R$ is the radius of the hemisphere, and $Y$ is the number of $\text{N}^{13}$
nuclei formed per second. Now, the number of $^{13}\text{N}$ nuclei formed in
time $dt$ would be

$$\gamma dt = \frac{J\rho A\sigma R}{2M} dt \quad (4)$$

The number of $^{13}\text{N}$ nuclei which decay in time $dt$, if $N$ exist at the be-
ingning of the interval, is $\lambda N dt$, where $\lambda$ is the disintegration
constant, so the change in the number of $^{13}\text{N}$ nuclei in existence in time
$dt$ is

$$dN = \gamma dt - \lambda N dt = (\gamma - \lambda N) dt \quad (5)$$

or, integrating and solving for $N$, and using the boundary conditions
that when $t = 0$, $N = 0$,

$$N = \frac{\gamma}{\lambda} (1 - e^{-\lambda t}) \quad (6)$$

So, after a bombardment time $T$, the number of $^{13}\text{N}$ nuclei present in the
sample will be

$$N_T = \frac{J\rho A\sigma R}{2M \lambda} (1 - e^{-\lambda T}) \quad (7)$$

or, the number of $^{13}\text{N}$ nuclei per ml of the irradiated nitrogen after
bombardment time $T$ will be, assuming a uniform distribution of the activ-
ity through the fluid,

$$n_T = \frac{N_T}{\frac{4}{3}\pi R^3} = \frac{3J\rho A\sigma R}{4\pi M\lambda R^2} (1 - e^{-\lambda T}) \quad (8)$$

Now consider the time at the end of the bombardment to be $t = 0$. At
this time, the $^{13}\text{N}$ will have reached a certain fraction of the concen-
tration it would have at $T = \infty$, and will decay exponentially at a
rate depending on its half-life. The observation of this decay begins at time $t_0$, which is the time it takes to carry the irradiated substance to the coincidence counting system and set it up. The decay is observed until time $t$. At the time $t=0$, there are $n_{13}^{13}$ nuclei per ml of nitrogen. At the time $t_0$, there are $n_{t_0} = n_{t} e^{-\lambda t_0}$ $N^{13}$ nuclei per ml, and at the end of time $t$, there are $n_t = n_{t} e^{-\lambda t}$ $N^{13}$ nuclei per ml. Then during the observation time $(t-t_0)$, the number of $N^{13}$ nuclei that will have decayed is $n_d = n_{t} (e^{-\lambda t_0} - e^{-\lambda t})$ $N^{13}$ nuclei per ml. The fraction of $n_d$ that is observed is $f$, which is determined in the calibration of the counting system with the standard Na$^{22}$ solution. The factor $f$ is defined as the ratio of the observed number of coincidences to the number occurring per unit volume, due to positron annihilation. So the number of coincidences that are observed during time $(t-t_0)$ is

$$n_D = f n_d = f n_t (e^{-\lambda t_0} - e^{-\lambda t})$$

(9)

or

$$n_t = \frac{n_D}{f (e^{-\lambda t_0} - e^{-\lambda t})}$$

(10)

from which, by substitution in equation (8),

$$\frac{3 J p A \sigma}{4 \pi M \lambda R^2} (1 - e^{-\lambda T}) = \frac{n_D}{f (e^{-\lambda t_0} - e^{-\lambda t})}$$

(11)

and, solving for $\sigma$,

$$\sigma = \frac{4 \pi M \lambda R^2 n_D}{3 J p A f (1-e^{-\lambda T})(e^{-\lambda t_0} - e^{-\lambda t})}$$

(12)

It should be mentioned at this point that $0.36\%$ of the liquid nitrogen
is $^{15}N$, but with such a small concentration, the correction in the

cross-section evaluation becomes negligible.$^4$

In the case of the Chlorine, the sample used for irradiation

is carbon tetrachloride. The only change in equation (12) then would

involve the number of nuclei per cm$^2$ in the hemispherical shell which

was first considered. If $M$ is the molecular weight of CCl$_4$, and

Avogadro's number is multiplied by 4, to indicate the presence of four

chlorine nuclei per molecule, and $\rho$ is the density of CCl$_4$, the cross-

section is given by

$$
\sigma = \frac{\pi M \lambda R^2 n_p}{3 J \rho A F (1.754)(1-e^{-\frac{\lambda}{\rho}})(e^{-\frac{\lambda t_0}{\rho}}-e^{-\frac{\lambda t}{\rho}})}
$$

(13)

where the factor $(1.754)$ in the denominator is the fraction of Cl$^{35}$
in the chlorine.$^4$

By similar considerations, the equation for the cross-section

for the $^{19}F$ reaction is given by

$$
\sigma = \frac{4 \pi M \lambda R^2 n_D}{9 J \rho A F (1.00)(1-e^{-\frac{\lambda}{\rho}})(e^{-\frac{\lambda t_0}{\rho}}-e^{-\frac{\lambda t}{\rho}})}
$$

(14)

B. Experimental Measurements.

The quantities in equations (12), (13), and (14) that must be
determined experimentally are $R$, $T$, $t_0$, $t$, $n_p$, $J$, and $\sigma$. No problem is
involved in the determination of $T$, $t_0$, and $t$. The value of $R$, of course,
is defined by the construction of the lucite hemisphere. Therefore, the
major part of the experimental measurement is concerned with the deter-
mination of $J$, $\sigma$, and $n_D$, which will now be discussed in that order.

(1) $J$, the Neutron Flux. In view of the geometry selected

for the activation of the target material, it was necessary to monitor

the neutron flux $J$ with a scintillation spectrometer. An anthracene
phosphor was used in conjunction with a 5819 photo-tube and conventional associated electronic circuits. The monitor was first calibrated absolutely by comparing its counting rate with that of an alpha-detecting proportional counter. The latter, which was constructed as part of a special tritium target chamber on the Cockcroft-Walton accelerator, used only for this calibration, was sensitive to the alpha particles from the $^{3}\text{T}_2(d,n)^{4}\text{He}$ reaction, subtending a solid angle of $3.534 \times 10^{-5} (\pm 1\%)$ of the total sphere. The anthracene monitor was placed 18 inches from the center of the target and perpendicular to the deuteron beam. The discrimination level of the monitor was adjusted so that only neutrons with energy above approximately 9 mev were counted. This setting was high enough to exclude most of the neutrons which were scattered from the floor and surrounding material, but yet allowed a reasonable counting rate of the neutrons concerned. The counting rate of a small Na$^{22}$ source was noted with the amplifier gain at an easily reproducible multiple of the original setting, and then later used as a reference point to reestablish the same gain and bias level whenever a bombardment was made. The total yield of neutrons per anthracene monitor count was found to be $4.08 \times 10^{-5} (\pm 2\%)$ before the experiment was performed, and $4.06 \times 10^{-5} (\pm 2\%)$ after the experiment was performed. The value used in the calculations, determined by a weighted average, is $4.07 \times 10^{-5} (\pm 1\%)$ neutrons/anthracene monitor count.

A correction must be made to the number above for the effect of the irradiated substance and its container. (Absorption, gamma rays from inelastic scattering of neutrons in the material, and possibly other effects.) This correction was determined by means of successive
measurements of neutron counting rates with the substance and container on the target and off. In the case of the nitrogen, the neutron flux must be increased by a factor of 14.0\% (±1\%), to compensate for these effects. The factor in the case of the carbon tetrachloride is 8.27\% (±0.3\%), and 16.2\% (±6\%) for the Kel-F polymer oil.

As mentioned before, the quantity J in the equation for the cross-section is the neutron flux per unit time. Unfortunately, it was impossible to keep this at a constant value during the irradiation, so an average value, determined by logging the monitor counting rate, was used. Fluctuations in the neutron flux from the average produce small effects on the net number of activated nuclei at the end of the irradiation, which can be corrected for from the log of the beam during irradiation. These corrections are discussed in Appendix II, with regards to the following types of fluctuations of the neutron flux:

1. Sinusoidal variations,
2. Gradual linear increases or decreases,
3. Abrupt terminations of the neutron flux at various stages in the irradiation interval, caused by spark discharges of the high voltage electrode of the Cockcroft-Walton. The correction to the average value of the neutron flux is only a few percent for all these cases, and has been made in the calculation of the cross-sections.

(2) Determination of f. The quantity f has been defined as the ratio of the observed number of coincidences to the number occurring per unit volume due to positron annihilation, and is a property of the counting system used, which is shown in Figure 2. The counters used were of the scintillation type with NaI crystals mounted on 5819 phototubes. The signals were amplified by linear amplifiers, and then passed
FIGURE 2 - THE COUNTING APPARATUS
into the coincidence circuit. The resolving time of the system was found to be about 0.3 microseconds.

The coincidence system was calibrated with a standard Na$_{22}$ solution, which had been calibrated absolutely previous to the experiment. (See Appendix I for the discussion of this calibration.) A calibrating solution for the system was made up of 1 ml of the absolutely calibrated solution to 7 ml of H$_2$O. The transferring of the solution was done by pipetting (delivery type), with an accuracy of ±0.2%. The strength of this calibrating solution at the time of the experiment, corrected for decay since its time of calibration (See Appendix I), was 1600 (±8%) disintegrations/second/ml. About 35-40 ml of the solution was poured into a test tube of the same diameter (1.125 inches) as that of the previously mentioned counting Dewar and this source was then centered in the counting system. As shown in Figure 2, Counter 1 was fixed, and Counter 2 could be revolved about the source to various angular positions. The source was centered in the system by turning up the amplifier bias of the movable channel (#2) to exclude background, and adjusting the position of the source so that the single channel counting rate was uniform regardless of the angular position of the counter. The lucite base, constructed to hold either the Na$_{22}$ test tube or the Dewar containing the irradiated substance, was then fixed in the system by means of paraffin.

When the Na$_{22}$ solution is placed and centered in the counting system, four sources contribute to the coincidences observed:

(1) accidentals, which can be calculated with a knowledge of the circuit resolving time and the single channel rates, (2) coincidences caused by
Compton scattering in the counters (significant in this case because of the close positions of the counters), (3) coincidences between the 1.28 mev and annihilation gamma rays, and (4) coincidences from the annihilation photons of the decaying positrons. The last group is the one that is to be examined. Since annihilation photons depart at 180° from each other, it is geometrically impossible for coincidences due to them to be observed with Counter 2 at a position less than 120° or greater than 240°. But at these positions the same number of coincidences of the other three groups is still observed. This is corroborated by the data shown in Figure 3, and it was concluded that the background counting rate could be found conveniently at either 112.5° or 247.5°, with the maximum rate at 180°. The rise in counting rates at 90° and 270° in Figure 3 is explained by an increase in Compton scattering, discussed further in Appendix IV.

When Counter 2 is at the 180° position, with the source at the center as shown in Figure 2, all the annihilation photon coincidences that are observed occur in a certain minimum volume of the solution, defined as the volume seen by both crystals in such a way that both annihilation photons from any single decay within the volume may be counted. If the test tube containing the Na\textsuperscript{22} source is so constructed and placed as to completely include this minimum volume, the number of coincidences observed will be a function of the number of coincidences occurring per unit volume, and will be independent of the kind of source. (This was concluded to be the case, when it was found that doubling or tripling the volume of the Na\textsuperscript{22} solution had no effect upon the net coincidence counting rate as long as the minimum volume was filled.) The net number of coincidences due to annihilation

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FIGURE 3

ANGULAR DISTRIBUTION OF COINCIDENCES

DOTTED LINES INDICATE BACKGROUND POSITIONS
photons (maximum rate minus background rate) is then divided by the known number occurring per unit volume, thus giving the factor $f$, which will be the same for any other irradiated substance under consideration. Hence, knowing the efficiency factor $f$ and the number of coincidences observed for the unknown radioactivity, one can compute its activity per unit volume. A correction must be made for the fact that the annihilation photons are absorbed by the $\text{Na}^{22}$ solution to a different extent than by either the nitrogen, the $\text{CCl}_4$, or the Kel-F oil, and a small uncertainty is also introduced by a slight variation of Compton scattering at various counter positions. (See Appendices III and IV.)

(3) $n_p$, The Net Number of Coincidences Observed. The factor $n_p$ is defined as the integrated number of coincidences due to annihilation photons that are observed during the time $(t - t_0)$. The coincidences observed in this case will be of the same sources listed above. Here again, only those due to positron annihilation are of interest, and can be determined by the background counting rates, as was done with the $\text{Na}^{22}$ solution. Thus, the counters are placed at $180^\circ$ and at background positions in alternate runs, and in each background run the integrated number of coincidences observed in time $(t - t_0)$ is determined. The normal room background (due to contamination in the room and cosmic ray background, causing both accidental coincidences and coincidences due to scattering), which is fairly constant, is then subtracted from the total background, and the result is normalized to the average neutron flux of the irradiation. The average room background, determined by overnight background counts, is 1.04 counts/min. In the
performance of the experiment, an attempt was made to keep the times $T$, $t_0$, and $t$ the same in all runs for each particular substance, in order to make this background meaningful without further calculation. In the case of the fluorine, a correction must be made for positions from decaying $^{34}$Cl nuclei. (The Kel-F oil is a polymer of trifluoro-chloroethylene.) This effect caused about twenty-four counts in this experiment, which were subtracted with the background to obtain $I_0$.

C. The Procedure of the Experiment.

Prior to each run for the measurement of a cross-section the counting system was calibrated with the $^{22}$Na calibrating source, by checking coincidence counting rates at the $180^\circ$ and the two background positions. The calibration of the neutron monitor was then checked with the small $^{22}$Na source, and the amplifier gain adjusted accordingly. The liquid to be irradiated was poured into the lucite container, and placed into position on the neutron source, as shown in Figure 1, with care being taken to keep bubbles from entering the hemisphere. Special precautions had to be taken in the case of the nitrogen, because of its low temperature. The lucite container had to be slowly cooled, and kept in a cylindrical Dewar flask during the experiment, to prevent excessive stress from the extreme temperature gradient. Precautions had to be taken to keep dry air in the container, to prevent moisture from condensing and freezing on the walls of the container tube, or possibly forming within the hemisphere and thus causing slight errors because of the change in volume of the nitrogen being irradiated. During the irradiation of the substance, the neutron flux was logged,
and the deuteron beam current was measured and kept approximately constant through the run. At the end of the bombardment, the liquid was transferred to the test tube Dewar, which was then placed in the counting system, and the coincidence counts were observed for the prescribed amount of time. Here again, special precautions had to be taken in the case of the nitrogen to prevent evaporation during the counting period. Weighing tests were made to determine the effects of evaporation, and it was found that when the irradiated nitrogen was poured into the Dewar (which had been cooled to liquid air temperature) and insulated from the outside air with glass wool, the rate of evaporation was about 0.7 gm/min. In the case of the nitrogen, the bombardment time \( T \) was 30 minutes, \( t_0 \) was 1.5 minutes, and \( t \) was 30 minutes. The bombardment time of the chlorine was 60 minutes, \( t_0 \) was 1.5 minutes, and \( t \) was 60 minutes. For the fluorine, \( T \) was 60 minutes, \( t_0 \) was 1.5 minutes, and \( t \) was 130.5 minutes.

D. Calculation of the Cross-sections.

The results of the calculations for the cross-sections are given as follows:

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Normalized Background (coinc. per anth. c/s)</th>
<th>( \frac{n_D}{J_F} )</th>
<th>( (\text{mb}) )</th>
<th>Uncertainty (( % ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{14}\text{N} )</td>
<td>0.62</td>
<td>( 1.7 \times 10^{-3} )</td>
<td>3.13</td>
<td>20</td>
</tr>
<tr>
<td>( ^{35}\text{Cl} )</td>
<td>3.4</td>
<td>( 1.5 \times 10^{-3} )</td>
<td>2.34</td>
<td>29</td>
</tr>
<tr>
<td>( ^{19}\text{F} )</td>
<td>16</td>
<td>( 3.4 \times 10^{-2} )</td>
<td>25.8</td>
<td>24</td>
</tr>
</tbody>
</table>
These values can be compared to those obtained in a different method by Paul and Clarke, who obtained 5.67 millibarns to accuracy of ±15% for the \(^{14}\text{N}(n,2n)^{13}\text{N}\) reaction, 3.47 millibarns to an accuracy of ±45% for the \(^{35}\text{Cl}(n,2n)^{34}\text{Cl}\) reaction, and 60.6 millibarns to an accuracy of ±30% for the \(^{19}\text{F}(n,2n)^{18}\text{F}\) reaction.  

E. Estimations of Errors. The uncertainties and errors in the experiment are estimated or calculated as follows:

(1) The determination of \(R\), the radius of the hemisphere, was made under two separate circumstances. The machine tool used to cut out the hemisphere from the lucite block was carefully measured, giving a radius of 3.02 \((± 0.5\%)\) cm. This value was adequate for the cases of the chlorine and the fluorine, since those irradiations took place at room temperature. However, cooling the lucite container to liquid air temperature may involve a change in \(R\). The temperature coefficients of linear expansion of lucite at the temperature of liquid air and room temperature are given as \(\frac{\Delta L}{L_{273.2}} = -8.58 \times 10^{-3}\) at 80°K, and \(\frac{\Delta L}{L_{273.2}} = +1.9 \times 10^{-3}\) at 300°K. Hence, the contraction involved in cooling the container to liquid air temperature should cause a radius contraction of about 1%. The radius of the hemisphere at liquid air temperature was also determined by a method of weighing. The lucite container was first weighed containing liquid air, and then empty, in order to determine the mass of liquid air filling the container. The radius of the hemisphere that would contain this amount of liquid air was determined to be 3.04 cm, with an uncertainty of ±1%.

(2) The uncertainties involved in the determination of \(n_p\)
are mainly to be found as statistical uncertainties. The average of the statistical deviations calculated for each run is \( \pm 6\% \) for the nitrogen, and \( \pm 20\% \) for the \( \text{CCl}_4 \). Only one run was made for the fluorine. Considering all of the runs that were made, the final statistical uncertainty for the nitrogen is \( \pm 3\% \), for the \( \text{CCl}_4 \) \( \pm 12\% \), and \( \pm 7\% \) for the fluorine. The difference in backgrounds due to scattering effects at the 180° and 112.5° positions is on the order of \( 1\% \), and within the statistical uncertainty of the background runs.

(3) There are several uncertainties involved in the calibration of the anthracene monitor. The determination of the solid angle subtended by the proportional counter is known to an accuracy of \( \pm 1\% \). The statistics of the calibration are accurate to \( \pm 1\% \). The results of the off-on determination of effects due to the irradiated substance and its container are known to an accuracy of \( \pm 1\% \). Effects due to sparking of the accelerator and possible change in amplifier gain or scalar bias put an uncertainty of \( \pm 2\% \) on the values for the neutron yield. Hence the total uncertainty of the value of \( J \) is \( \pm 5\% \).

(4) The greatest uncertainty in the determination of \( f \) is involved in the accuracy of the absolute calibration of the \( \text{Na}^{22} \) solution, which is known to an accuracy of \( \pm 8\% \). The statistics involved in the calibration of the coincidence equipment are usually on the order of \( \pm 1\% \), including the effect of a slight change in Compton scattering as Counter 2 is moved from 180° to 112.5°. (See Appendix IV) The corrections due to differences of absorption of gamma rays between the \( \text{Na}^{22} \), nitrogen, \( \text{CCl}_4 \), and Kel-F solutions are known to an accuracy of less than \( 1\% \). Shifts in the electronics during runs may cause
uncertainties on the order of $\pm 1\%$. The total uncertainty, then, in the determination of $f$ is taken to be $10\%$.

(5) Other miscellaneous effects causing uncertainties are involved in connection with the irradiated substance. A small amount of liquid is above the target during irradiation, which may cause a small uncertainty. Evaporation of liquid during the experiment may cause uncertainties of less than $1\%$ in the determination of the integrated counting rates. Ice falling into the irradiated nitrogen could possibly cause uncertainty, though none was noticed. The amount of irradiated liquid poured into the counting flask may differ slightly from one run to another, which would cause a very minor uncertainty. The finite size of the neutron source would cause some uncertainty on the order of $1\%$. Also, some pair production results from the 3.3 mev and 2.1 mev gamma rays in the C$^{34}$ decay, causing about 0.5 coincidence counts during the observation time ($t - t_0$), which is a small effect. (All other possible competing reactions were considered, and found to have no effect.)

The effects of all these uncertainties upon the calculated value of the cross-section can be seen by examining equations (12), (13) and (14). The experimental uncertainties introduced are $\pm 2\%$ for $R^2$, $\pm 5\%$ for $J$, $\pm 10\%$ for $f$, and the statistical uncertainty in $n_D$. (The error in $n_D$ also includes a certain amount of systematic error in the background determination.) Therefore the uncertainty in any calculated cross-section will be $\pm (17\% + \text{statistical uncertainty of } n_D)$. 

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APPENDIX I - ABSOLUTE CALIBRATION OF THE Na$^{22}$ SOURCE

The absolute calibration of the Na$^{22}$ source was done by two standard beta-counting techniques, for comparison. It was first done using conditions of "saturation backscattering", in terms of a standard RaD+E source, which was calibrated by the National Bureau of Standards. It was then done with a method of "defined geometries", using a source mounted on a very thin backing to reduce backscattering to negligible amounts. The two values thus obtained were averaged for the final result.

The apparatus used for the calibration with the standard RaD+E source is shown in Figure 4. The G-M tube was a Tracerlab model TGC2, end window type, with a 1.9 mg/cm$^2$ window. It was used with a model 165 combination scaling unit and power supply. Its operating voltage, determined from a plateau curve, was 1250 volts.

The frame shown in Figure 4 was of lucite, as were the supporting removable plates for the source and absorbers. The absorbers were composed of various thicknesses of aluminum foil. They were placed at about 1/3 the distance from the counter window to the source, to minimize scattering of particles into the counter. An aluminum absorber of thickness 2.65 mg/cm$^2$ was used in all measurements, to filter out any alpha rays from the Po$^{210}$ in the RaD+E source. This is denoted as the zero absorber thickness.

The standard RaD+E source #3078* consisted of 0.876 milli-

* Provided through the kind cooperation of Dr. W. J. Wingo of the M. D. Anderson Hospital for Cancer Research, Houston, Texas.
grams of lead electrolytically deposited as PbO₂ on a silver disk 1/16 inch thick faced with a layer of palladium 0.002 inch thick. The disintegration rate of the standard was computed by the National Bureau of Standards to be 532 disintegrations per second on September 9, 1949, with an overall uncertainty of ± 3%. The silver disk was encased in lucite supports as shown, with the source 4.395 cm from the counter window.

A silver disc of approximately the same size as that of the standard source was used for each Na₂²⁡ source, in order to insure saturation backscattering. A ring of Canada balsam was painted on the top face of each disc, to confine the area of the Na₂²⁡ solution to approximately the area of the RaD+3 standard. Each Na₂²⁡ source consisted of one drop of the solution to be calibrated, and enough of a very dilute detergent solution to fill out the designated area. The Na₂²⁡ drop was to an accuracy of ± 3% with a one milliliter capacity pipette (delivery type), calibrated to 1/100 milliliter. The source was allowed to evaporate overnight, the slow evaporation and the detergent solution serving to form a uniform deposit of the salt. Three Na₂²⁡ sources were made, and their counting rates compared. The one with a counting rate nearest the average was chosen for the calibration, which was ultimately calculated for the average source strength. The three sources were prepared as shown in Table 2.

<table>
<thead>
<tr>
<th>TABLE 2</th>
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<tbody>
<tr>
<td>Source</td>
</tr>
<tr>
<td>1</td>
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<tr>
<td>2</td>
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<td>3</td>
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<td></td>
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</tbody>
</table>

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The net counting rates of the Na$^{22}$ source and the standard were then observed as a function of the thickness of absorber. The absorption curves were plotted, and extrapolated to zero total absorber. (See Figure 5.) The net counting rate of the Na$^{22}$ source was then divided by that of the standard (both rates extrapolated to zero total absorber) and the quotient was multiplied by the disintegration rate of the standard to give the disintegration rate of the deposit of Na$^{22}$. Appropriate corrections were applied for decay of the standard since calibration$^6$, and for self-scattering in the RaD+E source. (The thickness of the source, of heavy molecular weight, causes self-scattering, which has been estimated to increase the counting rate of the source by a factor of 8%.$^6$)

The counting rates for various absorber thicknesses are shown in Table 3.

<table>
<thead>
<tr>
<th>Absorber №</th>
<th>Absorber thickness (mg/cm$^2$)</th>
<th>Net Na$^{22}$ rate (c/sec)</th>
<th>Net RaD+E rate (c/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.00</td>
<td>25.1</td>
<td>14.5</td>
</tr>
<tr>
<td>1</td>
<td>3.46</td>
<td>22.2</td>
<td>13.2</td>
</tr>
<tr>
<td>2</td>
<td>6.92</td>
<td>20.0</td>
<td>12.4</td>
</tr>
<tr>
<td>3</td>
<td>10.4</td>
<td>18.1</td>
<td>11.6</td>
</tr>
<tr>
<td>4</td>
<td>15.0</td>
<td>15.7</td>
<td>10.7</td>
</tr>
<tr>
<td>5</td>
<td>24.1</td>
<td>11.9</td>
<td>9.00</td>
</tr>
<tr>
<td>6</td>
<td>49.6</td>
<td>5.52</td>
<td>6.08</td>
</tr>
<tr>
<td>7</td>
<td>74.9</td>
<td>2.32</td>
<td>4.22</td>
</tr>
<tr>
<td>8</td>
<td>103</td>
<td>1.17</td>
<td>3.03</td>
</tr>
</tbody>
</table>

The average room background during the experiment was 1.003 c/sec. The maximum gamma background for the Na$^{22}$ source, due to 1.28 mev radiation and annihilation photons, was 0.632 c/sec, determined with enough aluminum absorber to completely cut out the positrons. Total background for the Na$^{22}$ data was then used as room background plus half maximum gamma.
background. (The exact gamma background was unknown, but the use of the half-maximum value could cause at most an error of only 1%)

The calculations are as follows:

Equivalent window thickness

Distance from source to window ........ 4.295 cm (±0.1%)
Density of air (26.6°C, 76.33 cm-Hg) ... 1.18 mg/cm³ (±1%)
Air absorption thickness ............ 5.08 mg/cm² (±1%)
Window thickness .................... 1.9 mg/cm² (±3%)
# 0 absorber thickness ............... 2.55 mg/cm² (±2%)

Equivalent Window Thickness .......... 9.63 mg/cm² (±2%)

Counting rates extrapolated to zero absorber (See Figure 5.)

Na²² .... 33.7 c/sec (±4%)    RaD-E .... 17.2 c/sec (±3%)

(8% correction for self-scattering in RaD+E leaves 15.8 c/sec.)

Relative strength of Na²² source ................. 2.13 (±7%)
Dis/sec of RaD-E standard (89.9% of original) .... 478 dis/sec (±3%)
Dis/sec of Na²² source #3 ................... 1020 dis/sec (±10%)

Strength of Na²² solution ................. 13,900 dis/sec/ml

Estimated uncertainty ................ ±14%

The second method of calibration is that of determining the source strength from the counting rate and the geometry of the apparatus. It involves fixing the geometry of the counting arrangement so that this factor is independent of the Geiger tube. This is done by means of a suitably machined orifice, placed between the source and the counter to collimate the counted particles into a defined geometric cone. It is
placed such that every particle in the cone will pass through the counting volume of the tube. Then by correcting the observed counting rate for absorption between source and counter, and considering the defined solid angle, the strength of the source can be determined.

The apparatus used is shown in Figure 6. The G-M tube, scalar and power supply, operating voltage, lucite frame, and absorbers are the same as before, with the exception that no "zero absorber" thickness was used.

The lucite collimator was made with a machined hole coaxial with the anode wire of the counter, such that the outermost particles from a point source would intersect the cathode about 2 cm from the counter window. The edge of the hole was bevelled to be parallel to the paths of the rays and thus reduce scattering.

The mounting for the source was made of a thin collodion film on a lucite ring. A drop of the collodion (commercial brand "New-Skin") was allowed to spread on the surface of water in a beaker, and the lucite ring brought up from underneath, such that a thin film was formed over the ring area. The film was found to be approximately 14.3 gm/cm², which is easily thin enough to make backscattering effects negligible.

Three sources were then prepared as before, with the exception that only one drop of the detergent solution was used. As before, their counting rates were averaged, the source with a counting rate nearest the average was used for the absorption curves, and the final calculation was made for the average source strength. The three sources were prepared as shown in Table 4.
### Table 4

<table>
<thead>
<tr>
<th>Source</th>
<th>Ml Na(^{22}) sol.</th>
<th>Net reg/min/ml</th>
<th>Average: 204 reg/m/ml</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.181</td>
<td>201</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.184</td>
<td>204</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0.160</td>
<td>208</td>
<td></td>
</tr>
</tbody>
</table>

Source Used: #2
Av/#2: 1.00 0.3%  

The counting rates for various absorber thicknesses are shown in Table 5.

### Table 5

<table>
<thead>
<tr>
<th>Absorber</th>
</tr>
</thead>
<tbody>
<tr>
<td>#</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Absorber thickness (mg/cm(^2))</th>
<th>Net c/sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>none</td>
<td>0.00</td>
</tr>
<tr>
<td>0</td>
<td>2.65</td>
</tr>
<tr>
<td>1</td>
<td>6.11</td>
</tr>
<tr>
<td>2</td>
<td>9.57</td>
</tr>
<tr>
<td>3</td>
<td>13.0</td>
</tr>
<tr>
<td>4</td>
<td>17.2</td>
</tr>
<tr>
<td>5</td>
<td>26.7</td>
</tr>
<tr>
<td>6</td>
<td>52.2</td>
</tr>
<tr>
<td>7</td>
<td>77.6</td>
</tr>
<tr>
<td>8</td>
<td>106</td>
</tr>
<tr>
<td></td>
<td>40.7</td>
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<td>36.5</td>
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<td>32.4</td>
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<td>23.9</td>
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<tr>
<td></td>
<td>18.5</td>
</tr>
<tr>
<td></td>
<td>8.33</td>
</tr>
<tr>
<td></td>
<td>3.57</td>
</tr>
<tr>
<td></td>
<td>1.30</td>
</tr>
</tbody>
</table>

The average room background during the absorption experiment was 1.24 c/sec. The average gamma background was determined by observing the counting rate with a 2.22 cm lead absorber in place. This thickness of lead would transmit 32\% of the 1.28 mev gammas, and only 2.4\% of the annihilation gammas formed at the lower face of the absorber. Then, assuming all the gamma background with zero absorber to be due to 1.28 mev gammas, the gamma background was calculated to be 0.99 c/sec, giving a total background of 2.23 c/sec. The absorption curve is shown in Figure 7. As in Figure 5, extreme curves were drawn, and an average value used.
The calculations are as follows:

**Equivalent Window Thickness**

- **Distance from source to counter window**... 4.03 cm (±0.1%)
- **Density of air (35.5°C, 75.92 cm-Hg)**... 1.18 mg/cm³ (±1%)
- **Absorption thickness of air**... 4.74 mg/cm² (±1%)
- **Counter window thickness**... 1.9 mg/cm² (±3%)
- **Equivalent window thickness**... 6.64 mg/cm² (±2%)

**Net counting rate, extrapolated to zero absorber**... 53.8 c/sec (±6%)

(See Figure 7)

**Geometry correction, made by means of Blachman's equations:**

\[
\begin{align*}
\alpha & = \text{distance from source to collimator} = 4.03 \text{ cm} \\
\beta & = \frac{b^2}{\alpha^2} = 0.087 \\
\gamma & = \frac{c^2}{\alpha^2} = 0.014 \\
\end{align*}
\]

**Blachman's equation:**

\[
G = 0.5 \left[ 1 - \frac{1}{(1+\beta)^{0.5}} - \frac{\beta \gamma}{(1+\beta)^{0.5}} \right] = 0.0202
\]

**Total activity of Na²² source #2**... 2660 c/sec (±6%)

**Amount of Na²² solution in source #2**... 0.184 ml (±1%)

**Strength of Na²² solution**... 14,500 c/sec/ml

**Estimated uncertainty**... ±7%

The final result was taken as the average of the two independent results:

**Average:** 14,200 c/sec/ml. **Uncertainty:** ±8%

The standard Na²² solution was calibrated on April 13, 1953. Its decay since that time makes a correction necessary to determine its strength during the performance of this experiment. This effect is
easily determined by means of a decay curve, shown in Figure 8. The half-life of Na$^{22}$ is taken to be 2.6 years. The date of this experiment was September 1, 1953, which is 0.384 years later than the calibration. Therefore, the strength of the Na$^{22}$ solution to be used for this experiment is 90.4% of the original value, or 12,800 dis/sec/ml (± 8%).
FIGURE 5 - $^{22}Na$ Absorption Compared with Rad+E
AV. 53.8 C/S

FIGURE 7 - Na^{22} ABSORPTION CURVE
It is desirable to calculate the percentage of error that would be caused by fluctuations of the deuteron beam of the Cockroft-Walton accelerator during the course of an irradiation. This error should be calculated with respect to the average value of the neutron flux, determined by logging the yield during the irradiation.

The case is first considered where the time variation of the neutron flux approximates a sine function. Equation (5) is referred to, expressing the number of radioactive nuclei in existence at the end of time dt, where \( N \) are in existence at the beginning of the interval: Now \( Y \), the yield of radioactive nuclei, is a function of the neutron flux \( J \), and if \( J \) is considered to be a sinusoidal function of the time, oscillating about an average value \( J_0 \) with an amplitude \( j \) and period \( 2\pi /\omega \), we have

\[
dN = K \left( J_0 + j \sin \omega t \right) dt - \lambda N dt
\]

or

\[
\frac{dN}{dt} + \lambda N = K \left( J_0 + j \sin \omega t \right)
\]

(14)

where \( K \) is a constant representing the fraction of the neutron flux that reacts with the substance being irradiated. Equation (14) is a simple linear equation that can be integrated and solved for \( N \), using the boundary conditions that when \( t = 0 \), \( N = 0 \) to determine the arbitrary
constant, giving

\[ N = \frac{KJ_0}{\lambda} \left(1 - e^{-\lambda t}\right) + \frac{Kj}{\lambda^2 + \omega^2} \left[ \omega e^{-\lambda t} + \lambda \sin \omega t - \omega \cos \omega t \right] \]  

(15)

the first term in equation (15) is recognized as equation (6), and the correction to be made for fluctuation of the beam lies in the second term of equation (15). Suppose that the neutron flux has an average value of \(10^7\) neutrons/sec, with a fluctuation of \(\pm 20\%\) in a 30 minute irradiation. Then the value of \(j\) would be \(2 \times 10^6\) neutrons/sec. Let the oscillation go through 1.25 cycles (2.5 \(\pi\) radians, which was found to be a reasonable number, corresponding quite well with actual conditions), with the bombardment ending when the neutron yield is at its maximum point. Then the value of the first term is found to be \(7.7 \times 10^9\) K, using the half-life for H\(^{13}\), and the value of the second term is \(0.17 \times 10^9\) K. Therefore, the correction involves a 2.3\% addition to the average value. If the case is considered where the oscillation undergoes 1.5 cycles (3 \(\pi\) radians) in the 30 minutes of bombardment, it is found that a 5\% correction must be made to the average value, which is about the maximum correction that must be made in any case. In each case, the value of \(\omega\) is determined by the relation \(\omega = n \cdot \pi / t\), where \(n\) is the number of radians the oscillation undergoes during bombardment time \(t\).

Equation (18) can be used with any of the irradiations of either of the three substances, with the corresponding values of \(t\) and \(\lambda\), and the proper values of \(J_0\) and \(j\) (which are determined from plotting the neutron flux as a function of irradiation time), with the restriction that the fluctuation be approximately sinusoidal.
The case is now considered where the beam starts out below its average value, and gradually increases by a factor of 30\% during the bombardment time $T$. Then the change in the number of radioactive nuclei present during time interval $dt$ is given by

$$dN = K \left[ J_0 + j \left( t - \frac{T}{2} \right) \right] dt - \lambda N dt$$

(16)

where $j$ is the slope of the line representing the variation of neutron flux with time.

Equation (16) can be re-written as

$$\frac{dN}{dt} + \lambda N = K \left[ J_0 + j \left( t - \frac{T}{2} \right) \right]$$

in the standard form of a simple linear equation. Integrating and solving for $N$, using boundary conditions that when $t = 0$, $N = 0$,

$$N = \frac{KJ_0}{\lambda} (1 - e^{-\lambda t}) + \frac{Kj}{\lambda^2} (\lambda t - 1 + e^{-\lambda t}) - \frac{KjT}{2\lambda} (1 - e^{-\lambda t})$$

(17)

Or, if the beam starts out high and gradually decreases,

$$N = \frac{KJ_0}{\lambda} (1 - e^{-\lambda t}) - \frac{Kj}{\lambda^2} (\lambda t - 1 + e^{-\lambda t}) + \frac{KjT}{2\lambda} (1 - e^{-\lambda t})$$

(18)

Now consider the case as mentioned where the beam increases by 30\% of its original value, during a 30 minute bombardment time. The first term, which is merely equation (6) again, is evaluated as $7.6 \times 10^9$ K neutrons/sec, and the third term is $0.988 \times 10^9$ K neutrons/sec. Hence

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the total correction, considering algebraic signs, amounts to 4.2\% in this case. Equation (18) can also be applied to any of the irradiations, with the appropriate values of \( \lambda \), \( t \), \( J_0 \), and \( j \). In the procedure of this experiment, the neutron flux was logged and plotted as a function of the irradiation time, and the appropriate correction made for each value of \( J \) in the determination of the cross-section.

The effect is now considered of a spark discharge of the accelerator during the irradiation, where the neutron flux suddenly drops to zero for a period of approximately 15 seconds, and is then brought back to normal. Equation (6) is referred to again, which was developed from equation (5), using the boundary conditions that when \( t = 0, N = 0 \). It is now assumed that when \( t = 0, N = N_0 \), considering the time that the beam is turned back on again to be the time \( t = 0 \). Then by integrating equation (5), and using these boundary conditions, the expression is obtained:

\[
N = \frac{KJ_0}{\lambda} (1 - e^{-\lambda t}) + N_0 e^{-\lambda t}
\]

(19)

Now suppose that the machine sparks at \( t = 28 \) minutes (1680 seconds), and is turned back on again at \( t = 28 \) min, 15 seconds. Then

\[
N_0 = \frac{KJ_0}{\lambda} (1 - e^{1680 \lambda}) e^{-15 \lambda}
\]
considering the decay during the 15 seconds that the beam was off, and it is found from equation (19) that at the end of 105 more seconds after the beam is turned on, which completes the 30 minute bombardment time, there are \(0.860 \frac{\text{KJ}_0}{\lambda}\) radioactive nuclei. If there had been no sparking, there would have been \(0.874 \frac{\text{KJ}_0}{\lambda}\) radioactive nuclei. Hence, the spark involved a 1.6% correction. If the spark had occurred earlier, the correction would have been even less. To take an extreme case, suppose that another spark occurs just 60 seconds after this previously considered spark. By the same method, it is found that at the end of the 30 minute bombardment time there are \(0.856 \frac{\text{KJ}_0}{\lambda}\) radioactive nuclei, as compared with \(0.874 \frac{\text{KJ}_0}{\lambda}\) radioactive nuclei with no sparks at all, which involves a correction of 2.1%.
In the calculation of \( f \), a correction becomes necessary to take into account the differences in absorption of gamma rays between the \( \text{Na}^{22} \) solution and the irradiated substance. Considering first the case of the nitrogen, suppose that a position decays at some point in the minimum counting volume, such that both annihilation photons could be counted. If the photons are produced at such a point that one must pass through a thickness \( x_1 \) of the nitrogen before going into Counter 1, and the other through a thickness \( x_2 \) before going into Counter 2, the probabilities that the photons will be counted are \( e^{-\mu_n x_1} \) and \( e^{-\mu_n x_2} \), respectively, and the probability that a coincidence will be observed between the two is \( e^{-\mu_n (x_1 + x_2)} \). Similarly, in the case of the \( \text{Na}^{22} \) solution it is \( e^{-\mu_s (x_1 + x_2)} \) for the carbon tetrachloride \( e^{-\mu_c (x_1 + x_2)} \) and \( e^{-\mu_f (x_1 + x_2)} \) for the Kel-F oil, where \( \mu_n, \mu_s, \mu_c, \) and \( \mu_f \) are the total absorption coefficients for annihilation photons in their respective mediums.

Then, making a comparison between the \( \text{Na}^{22} \) solution and the other three, considering the same photons originating at the same point, it is seen that

\[
\frac{N_s}{N_n} = e^{-X (\mu_s - \mu_n)}, \quad \frac{N_s}{N_c} = e^{-X (\mu_s - \mu_c)} , \quad \text{and} \quad \frac{N_s}{N_f} = e^{-X (\mu_s - \mu_f)}
\]

where \( X \) is the total distance through the liquid. The values for the absorption coefficients are \( \mu_s = 0.108 \text{ cm}^{-1}, \mu_n = 0.0697 \text{ cm}^{-1}, \mu_c = 0.134 \text{ cm}^{-1}, \) and \( \mu_f = 0.161 \text{ cm}^{-1}. \) The value for the total distance \( X \) is 2.86 cm. Hence the relative amounts of absorption for the \( \text{Na}^{22} \) solution as compared to the other three are

\[
\frac{N_s}{N_n} = 0.896, \quad \frac{N_s}{N_c} = 1.077, \quad \text{and} \quad \frac{N_s}{N_f} = 1.167.
\]
Then by means of these corrections, the number of annihilation photons produced in the $^{22}$Na solution that would be seen by the counters if the absorptivity of the $^{22}$Na solution were the same as that of the irradiated substance under consideration can be determined for each factor $f$. 
A small uncertainty is caused in the determination of counting backgrounds, because of the slight variation in Compton scattering with various angular positions of the counters. With a Cs\(^{137}\) source at the center, the following coincidence counting rate is observed for various angular positions of Counter 2:

<table>
<thead>
<tr>
<th>Angle</th>
<th>(\mu/\text{min})</th>
</tr>
</thead>
<tbody>
<tr>
<td>180°</td>
<td>28.0</td>
</tr>
<tr>
<td>210°</td>
<td>28.1</td>
</tr>
<tr>
<td>240°</td>
<td>29.2</td>
</tr>
<tr>
<td>247.5°</td>
<td>30.3</td>
</tr>
<tr>
<td>270°</td>
<td>35.2</td>
</tr>
</tbody>
</table>

The number of accidental coincidences occurring is 12.9 reg/min, determined from the constant single channel counting rates, so the net numbers of coincidences due to Compton scattering at 180° and 247.5° are 15.1 reg/min and 17.4 reg/min respectively. This would indicate a decrease in Compton scattering effect of 13\% in changing from the background position to the 180° position. Now, when the Na\(^{22}\) source is in place, the counting rate at background is approximately 42 reg/10 min. About two of these registers are due to accidentals, and it is found through calculating the efficiencies of the counters by means of the standard Na\(^{22}\) solution that about 23 reg/10 min. will be due to real coincidences between 1.28 mev and 0.51 mev gamma rays. Therefore, about eleven of the registers are due to Compton scattering, and the decrease of background in changing positions from 247.5° to 180° would be about
1.4 registers over the ten minute counting period. This would amount to about a 3\% decrease in background, then, as the counter is moved from 247.5° to 180°.
ACKNOWLEDGMENTS

The author is deeply indebted to Dr. C. M. Class, who suggested the problem. His many valuable discussions, aid, and advice were indispensable to the completion of the project. Appreciation is also expressed to Messrs. Van der Henst and De Vries of the Physics Shop for their cooperation and advice in the construction of the equipment.
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8 - B. Burtt, Nucleonics 5, No. 2, 28 (1949).
