MASSACHUSETTS INSTITUTE OF TECHNOLOGY

Laboratory for Nuclear Science

PROGRESS REPORT

February 28, 1955
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MASSACHUSETTS INSTITUTE OF TECHNOLOGY
Laboratory for Nuclear Science
Cambridge 39, Massachusetts

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PROGRESS REPORT

February 28, 1955

The data and the results which are presented in this report should not be published elsewhere without prior consultation with the Laboratory for Nuclear Science.

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This is the thirty-sixth progress report of the Laboratory for Nuclear Science at the Massachusetts Institute of Technology. Progress during the period of November 30, 1954 through February 28, 1955 is reported.

**TABLE OF CONTENTS**

<table>
<thead>
<tr>
<th>Group</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemistry of the Fission Elements Group</td>
<td>1</td>
</tr>
<tr>
<td>Nuclear Chemistry (Inorganic) Group</td>
<td>9</td>
</tr>
<tr>
<td>Nuclear Chemistry (Organic) Group</td>
<td>17</td>
</tr>
<tr>
<td>Cosmic Ray Group</td>
<td>21</td>
</tr>
<tr>
<td>Elementary Particle Scattering Group</td>
<td>27</td>
</tr>
<tr>
<td>Neutron Physics Group</td>
<td>33</td>
</tr>
<tr>
<td>ONR Generator Group</td>
<td>43</td>
</tr>
<tr>
<td>Radioactivity Group</td>
<td>51</td>
</tr>
<tr>
<td>Cyclotron Group</td>
<td>53</td>
</tr>
<tr>
<td>Synchrotron Group</td>
<td>55</td>
</tr>
<tr>
<td>Theoretical Group</td>
<td>61</td>
</tr>
<tr>
<td>Personnel Listing</td>
<td>67</td>
</tr>
</tbody>
</table>

Reprints of published papers listed herein are available in limited numbers upon request from the Laboratory for Nuclear Science.
Chemistry of the Fission Elements Group

I. STUDIES OF COMPLEX IONS

A. Halide Complexes of Indium

The ultraviolet spectrophotometric study of bromide complexing of indium in aqueous solution has been continued.

The spectra for indium in the presence of a slight excess of bromide for wavelengths 226-240 m\(\mu\) at an ionic strength of 4.00 were determined. The data were interpreted by a slightly modified McConnell-Davidson method\(^1\) for the determination of the second stability constant, that is, \(\text{InBr}^{++} + \text{Br}^- \rightarrow \text{InBr}_2^+\), and \(K_2 = \frac{(\text{InBr}_2^+)}{(\text{InBr}^{++})(\text{Br}^-)}\). \(K_2\) was found to be 18.7 ± 0.5.

From a logarithmic plot similar to that used by Kingery and Hume\(^2\), the stability constant for the reaction \(\text{InBr}^{++-n} + p\text{Br}^- \rightarrow \text{InBr}^{++-n-p}\) was determined. The stability constant, \(K_{n+p}\), equals \((\text{InBr}^{++-n-p}_{n+p})/((\text{InBr}^{++-n}_{n+p})(\text{Br}^-)^p\), where \(n+p\) is the maximum number of ligands of bromide on indium and \(n\) is the number of ligands of bromide on the next lower complex. In the wavelength region 235-244 m\(\mu\) and 260-268 m\(\mu\) and an ionic strength of 4.00, \(K_{n+p} = 6.7 \pm 0.6\) and \(p = 1.12 \pm 0.04\).

Previous studies by the method of continuous variations indicated that a four to one complex is the highest complex. In recent work at an ionic strength of 6.80 M and reacting constituents of 1.00 M, a 4.4 to 1 complex is indicated. It is difficult to evaluate this evidence. However, if \(K_{n+p}\) is assumed to be \(K_4\), the data do not yield a constant value of \(K_3\). It is possible that the method of continuous variations is not valid under the conditions of these experiments. This possibility will be explored further in order to interpret the data already obtained.

(E. A. Burns)

B. Ferric Thioglycolate Complexes

Spectrophotometric studies have been continued on the autoreduction of the ferric thioglycolate complex formed in alkaline media. The overall reaction is:
Chemistry of the Fission Elements Group

\[ 2\text{SCH}_2\text{CO}_2^- + 2\text{Fe(III) complex} \rightarrow 2\text{Fe(II) complex} + \text{O}_2\text{CCH}_2\text{SSCH}_2\text{CO}_2^- \]

The course of the reaction has been followed by measuring the absorbancy of the ferric complex at 530 m\(\mu\) as a function of time. The concentration of thioglycolate in the solution was in excess so that its concentration remained constant during a run. An ammonia-ammonium nitrate buffer was used. The rate can be described by the equation:

\[ \frac{-dA}{dt} = (k_1 + \frac{k_2}{\text{TGA}}) A^2. \]

\(A\) represents the absorbancy at 530 m\(\mu\) and \(\text{TGA}\) represents the thioglycolate concentration. The rate constant, \(k_1\), is independent of the pH in the range investigated (pH 8.4 - 10.1). The rate constant, \(k_2\), is a function of pH. The data are being interpreted to obtain a physical picture of the reaction mechanism.

(L. Newman)

II. ELECTRODEPOSITION BEHAVIOR OF TRACES

To study the fixed potential deposition behavior of nickel in solutions too dilute to follow chemically, radioactive tracers will be used. To best utilize the tracers, we are building a differential counting rate meter. This device is comprised of two counting rate meters with opposed outputs. One rate meter is connected to a Geiger dip-type counter which measures the radioactivity in the electrolytic solution; the other rate meter is connected to a Geiger counter which monitors a reference standard of the same isotope. The output from the differential counting rate meter can thus be made to indicate directly the removal of radioactive nickel from the electrolytic solution, independent of the decay of the isotope.

The differential counting rate meter consists of two Geiger counter triggered "single shot" multivibrators which drive condenser step chargers. The voltages thus developed across the two condensers are fed to the opposing grids of a vacuum tube voltmeter. The voltmeter output is taken via a cathode follower to a recording potentiometer. This device will make it possible to follow continuously the progress of the electrolysis with respect to rate of deposition and completeness of the separation. (C. F. Morrison, Jr.)
III. POTENTIOMETRIC MEASUREMENTS IN DILUTE SOLUTIONS

The study of the sensitivity of the bromine-bromide potentiometric endpoint by means of a coulometric generation of sub-micro quantities of bromine has been continued.

The differences between the results obtained with a Leeds and Northrup pH meter (Cat. No. 7664), a Rubicon potentiometer (Cat. No. 8702), and a Beckman Model G pH meter reported by Mr. W. C. Purdy of this laboratory have been confirmed. However, the earlier conception that the blank wave was due to the stripping off of dissolved or adsorbed gases on the indicator electrode appears to be in error. If a solution is diluted with supporting electrolyte (0.200 M sodium bromide and 0.100 M sulfuric acid) after sufficient bromine has been generated to obtain theoretical behavior according to the Nernst equation the dilution curve does not retreat according to the equation, but instead retraces rather well the generation curve. One would think that the blank could be due to titration of an impurity from this information. However, from the previously reported information of the constancy of the blank with varying concentrations of sodium bromide and sulfuric acid, it is evident that this is not the case, and the blank must be due to some inherent characteristic of the indicator electrode.

At the present time, it is felt that the blank may be due to a combination of effects. First, the initial potential is a "mixed" potential of the adsorbed hydrogen and dissolved oxygen couples which is variable (within about sixty millivolts). Second, there is a certain limiting concentration of bromine which is necessary to assume control of the indicating electrode reaction. Finally, Professor C. E. Bricker of Princeton University has discussed some of his work with J. K. Lee and R. N. Adams which indicates that electrode films may also be a factor. Experiments designed to test these ideas are now under way. (E. A. Burns)

IV. COULOMETRY

Mercury was dissolved from an amalgamated silver wire anode at 100% current efficiency. Potentiometric titration curves were obtained on a recording potentiometer using a similar indicator electrode versus a reference electrode. The accuracy in all of these titrations was comparable to that reported for coulometric
Chemistry of the Fission Elements Group

titrations of halides with silver and the sensitivity at very low concentrations was better than for analogous silver titrations. The coprecipitation errors encountered in argentometric titrations of halide mixtures are also present in this method, whereas the usual adsorption errors in the titration of iodide alone were not observed. Further studies are being carried out to extend the use of generated mercurous and mercuric ions for the determination of other species.
(E. P. Przybylowicz)

V. NON-AQUEOUS TITRATIONS

The behavior of the glass electrode and the magnitude of the sodium ion error have been extensively studied in aqueous media. However, few investigators have examined the deviations caused by sodium ion when the glass electrode is used to measure pH in non-aqueous solutions of high alkalinity. The present study is an attempt to measure that error by following an acid-base reaction with two different electrode pairs. The reference electrode, in both cases, is composed of a saturated solution of tetramethyl ammonium chloride in absolute alcohol in contact with a mixture of mercury and calomel. One indicator electrode is the glass electrode and the other is a hydrogen electrode. Any deviation in potential of the glass electrode, due to sodium ion, can be determined by comparing the glass electrode potential with that of the hydrogen electrode, which is not affected by sodium ion.

Preliminary runs, with these electrode pairs, have been made in aqueous solutions in order to establish the qualitative relationship between the general behavior of the two pairs of electrodes. Also, some titrations of glacial acetic acid in absolute alcohol were performed with sodium ethylate as the titrant. However, not enough data have been gathered to present any definite conclusions at this time.
(R. C. DeGeiso)

VI. AUTOMATIC THERMOMETRIC TITRATIONS

The recording of the unbalance potential of a Wheatstone bridge circuit, containing a thermistor as the active arm, indicates continuously the temperature change of the solution during the automatic titration process. The calibration of the unbalance potential in terms of temperature change is not necessary for the de-
tection of endpoints, and in previous work only the order of magnitude of the changes have been estimated. However, in glacial acetic acid (solvent), the shapes of the titration curves differ significantly depending upon the substance being titrated, and endpoints have resulted from both positive and negative temperature changes. In order to tabulate and correlate the temperature-change data, the thermistor bridge has been rebuilt to permit a direct calibration of the unbalance potential in terms of temperature change. This new bridge provides a dynamic measurement of temperature change, that is, does not require a rebalancing of the bridge and subsequent interpolation of the temperature between two points (at balance position) on the potential axis of the recording. In order to convert temperature-change data into heats of reaction, a simple electrical heater circuit has been built to measure the heat capacities of the solutions titrated.

The water content of the acetic acid solvent has been reported previously as an important factor affecting the heats of mixing during a titration. Subsequent work in anhydrous acetic acid has shown that the temperature rise accompanying the titration of sodium acetate is two to three times as great as that obtained in glacial acetic acid (approximately 0.3% water present in glacial acetic acid). Work is planned with Karl Fischer reagent which will establish more exactly the water contents of the solutions being titrated. In this way the influence of water upon the thermometric endpoint will be defined more clearly. (H. J. Keily)

VII. NEPHELOMETRIC STUDIES

During the recent investigation of a nephelometric procedure for the determination of the sulfate impurity in reagent-grade salts, two distinctly different calibration curves were obtained with identical solutions depending upon the method of addition of the barium chloride reagent. An investigation has been undertaken to examine the precipitation conditions which influence the development of the turbidities. It has been found that the spectra obtained by light absorption measurements at various wavelengths in the ultraviolet region are not changed by the level of the sulfate present in the solutions. However, precipitation by solid barium chloride and by a solution of barium chloride resulted in distinctly different spectra. These spectra can probably be interpreted in terms of differences in the particle size distribution of the barium sulfate. Further work is planned to define the factors influencing the precipitation process. (H. J. Keily)
Chemistry of the Fission Elements Group

VIII. RADIOMETRIC ANALYSIS

Manganese has been determined in biological materials by activation analysis. The procedure involves the irradiation of the ashed biological sample in the MIT cyclotron (flux $1.2 \times 10^8$ neutrons/cm$^2$/sec.) for 12 hours. Following irradiation the sample is dissolved in nitric acid and a known amount of inactive manganese carrier is added. A manganese separation is performed and the chemical yield of the separation is determined colorimetrically. The sample is then counted and the manganese content is calculated by comparison to a standard manganese sample that has been put through the same procedure as the unknown.

By this method, the manganese contents of beef liver and spleen, chicken liver, and a bacterial culture have been determined. The results compare favorably with those obtained by the standard periodate method for manganese. The activation method has been found useful for submicrogram quantities of manganese due to the elimination of the usual chemical interferences and the added sensitivity of radioactivity measurement. Plans are under way to extend the method to amounts below the colorimetric limit by using the higher neutron flux at Brookhaven National Laboratory. (W. C. Purdy)

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ADDRESSES


-6-
Chemistry of the Fission Elements Group


PUBLICATIONS


D. N. Hume
L. B. Rogers
The Extraction of Ga(III) from Aqueous HCl by Organic Solvents

Studies on the extraction of Ga(III) from aqueous HCl by organic solvents has been extended to diisopropyl and dihexyl ether. Contrary to the metal-ion concentration dependence of the distribution constant, E, reported for the system Ga(III) - HCl - β,β'-dichlorodiethyl ether, the diethyl, diisopropyl, and dihexyl ethers showed no such effect. In these systems E remains constant (E = 50, 40, and 0.7, at 6 N, 6 N, and 9 N HCl respectively, with aqueous phase gallium concentrations of $10^{-3} \text{F}$ to carrier free ($< 10^{-8} \text{F}$) gallium tracer. In the gallium concentration region $10^{-3} \text{F}$ to $10^{-1} \text{F}$, the Nernst distribution law is no longer obeyed, E increasing up to 5 or 6 fold at the higher concentration.

In the diethyl- and diisopropyl-ether systems, carrier-free gallium shows a maximum distribution constant between 6 and 8N HCl. No such maximum is observed in the dihexyl ether extractions, E increasing continuously up to 12N hydrochloric acid.

The effect of the addition of small amounts of ClO$_4^-$ and NO$_3^-$ on the distribution constant E of Ga(III) between β,β'-dichlorodiethyl ether and HCl has been investigated more exhaustively for the system: carrier-free Ga(III) tracer - 3.32N HCl - β,β'-dichlorodiethyl ether. It has been found that the distribution constant falls from ~5.2 in a ClO$_4^-$-free system to ~0.15 at $5 \times 10^{-2} \text{F}$ aqueous ClO$_4^-$. A system 3.32N in HCl and $10^{-4} \text{F}$ in [ClO$_4^-$]$_a$ shows a depression of E from ~5.2 (for the ClO$_4^-$-free system) to 2.4.

It appears that the interpretation of these data involve the extraction of HCl, HClO$_4$, and HGaCl$_4$ from the aqueous to the organic phase and the dissociation of these acids in the ether. A series of conductivity measurements of aliquots of β,β'-dichlorodiethyl ether equilibrated with aqueous HCl, HClO$_4$, and HNO$_3$ shows the conductance order HCl < HNO$_3$ < HClO$_4$.

Titrimetric measurements for total acid extracted by the organic layer from aqueous HCl, HClO$_4$, and HNO$_3$ gives the solubility order HCl < HClO$_4$ < HNO$_3$, in
Nuclear Chemistry (Inorganic) Group

the aqueous phase concentration region 0.1 - 12N, 0.1 - 5.5N, and 0.1 - 5N respectively.

These studies are being extended to considerations of relative acid strengths as a function of dielectric constant of the organic phase, generalized acid-base phenomena, and hydrogen bonding as an interpretation of the observed extraction data. (E. Rudzitis, R. H. Herber and J. W. Irvine, Jr.)

II. PHYSICAL-CHEMICAL BACKGROUND FOR RADIOCHEMICAL SEPARATIONS

Quantitative Treatment of the Strengths of Strong Acids

Strong mineral acids are used in many radiochemical operations, and a unified physico-chemical treatment of the chemical factors underlying these and similar processes requires a sharpening of the quantitative treatment of the ionization properties and effective acidity of solutions of strong acids varying up to large concentration in mixtures with water.

One of the most useful treatments is that due to Hammett\(^1\) who has extended the classical indicator method to determine the function \(H_0\) using indicator with one positive charge in the acid form \(BH^+\) and with zero charge in the basic form \(B\). The Hammett function \(H_0\) is given\(^1\)

\[
H_0 = -\log a_{H^+} - \log_{10} \frac{f_B/f_{BH^+}}{f_{BH^+}} = \log \frac{(B)}{(BH^+)} + pK_{BH^+} \quad (1)
\]

where \(a_{H^+}\) is the activity of free protons, \(f_{BH^+}\) and \(f_B\) are the activity coefficients of the two indicator forms, and \(pK_{BH^+}\) is the negative logarithm of the indicator acid-ionization constant. Data are available\(^1\) for \(H_2SO_4\) from 0 to 100\%, and on into \(H_2SO_4-\)\(SO_3\) mixtures\(^2\) using substituted nitrobenzenes as indicators. The \(H_0\) of 100\% \(H_2SO_4\) is -10.6, corresponding to a proton activity \(10^{10.6}\) higher than that in 1\(M\) \(H_2SO_4\) solution in water. Sulfuric acid is thus a superacid, as are the still stronger acids \(HClO_4\) and the polysulfuric acids \(H_2SnO_{3n+1}\). For aqueous solutions of \(H_2SO_4\) from 10\% (1\(M\)) up to 95\% (17.8\(M\)), \(H_0\) can be correlated with the molarity \(M\) as:

\[
H_0 = 0.5M - 0.3 \quad (2)
\]

with an average deviation of \(\pm 0.08\) log units.

The present study has been concerned first with removing the restrictions on charge-type of the indicator. Hammett\(^1\) has shown that the activity coefficient...
ratio \( f_B/f_{BH^+} \) is independent of concentration of \( \text{H}_2\text{SO}_4 \) above 85%. Comparison of the Michaelis acidity function \( \bar{G} \), dependent on the activity coefficients involved in semiquinone formation of multipositively charged dyestuffs, indicates that the activity coefficients for typical components have become independent of acid concentration above 4 - 6M \( \text{H}_2\text{SO}_4 \). Further support is available for the concentration independence in the study \( \text{H}_+ \) with picric acid as indicator. Thus there can be established the relation, good above 4 - 6M \( \text{H}_2\text{SO}_4 \):

\[
\text{H}_x - \text{H}_0 = C_x \quad (M>6)
\]

where \( \text{H}_x \) is a Hammett generalized function based on indicator of charge +x in the basic form, and \( C_x \) is the corresponding constant, equal to \( \log (f_{BH^+} + f_x)/f_Bf_x + 1 \), accessible by curve-fitting to the \( \text{H}_0 \) curve. This justifies the treatment of Lewis and Bigeleisen\(^5\) of their data with the indicator dibromodinitrofluorescein, which probably measures \( \text{H}_{+3} \) but was fitted to the \( \text{H}_0 \) function. Lewis and Bigeleisen went further to correlate the vapor pressure of \( \text{SO}_3 \) \( P_{\text{SO}_3} \) to the \( \text{H}_0 \) scale:

\[
\text{H}_0 = -\log P_{\text{SO}_3} - 14.56
\]

Lewis and Bigeleisen state that \( \log a_{H^+} \) should fall with increasing \( \text{SO}_3 \) content of \( \text{H}_2\text{O} - \text{SO}_3 \) mixtures with a maximum just short of 50 mole % \( \text{SO}_3 \). This seems unreasonable to us, and the data have been reinterpreted showing parallelism between \( -\log a_{H^+} \) and \( -\log P_{\text{SO}_3} \), based on acidification by increasing the chain length in the typical polysulfuric acid, changing \( \text{H}_2\text{O} (\text{SO}_3)_n \) to \( \text{H}_2\text{O} (\text{SO}_3)_{n+1} \). The typical ionization reaction will be:

\[
\text{H}_2\text{O} (\text{SO}_3)_n + \text{H}_2\text{O} (\text{SO}_3)_{n-1} = \text{H}_3\text{O} (\text{SO}_3)_{n-1}^+ + \text{HO} (\text{SO}_3)_n^-
\]

with the successive polysulfuric acids representing substantially stronger acids as the value of \( n \) goes up.

The increase in fugacity of \( \text{SO}_3 \) in the system should have the same effect on an indicator as the increase in fugacity of the free proton \( H^+ \).

The treatment of the \( \text{H}_2\text{SO}_4 - \text{SO}_3 \) system is the subject of a paper appearing in issue 1 of the new Journal of Inorganic and Nuclear Chemistry.

Attention is also being given to the strength of other mineral acids in concentrated aqueous media. (C. D. Coryell and R. C. Fix)
III. GENERAL RADIOCHEMICAL STUDIES

A. Search for Pb\textsuperscript{206}

Investigations to observe radioactive Pb\textsuperscript{206} are presently aimed at detecting a very short (minutes \rightarrow months) or very long (years \rightarrow geologic) half-life.

Lead foil was bombarded by deuterons in the MIT cyclotron, dissolved in 6N HNO\textsubscript{3}, 5 mgm of Bi carrier added, and the Bi activity purified from lead by precipitation of Pb(NO\textsubscript{3})\textsubscript{2} with fuming nitric acid. The Bi\textsuperscript{4+3} in 0.5N HCl is then carried onto a Dowex-1 (7 1/2\% DVB) resin column. Milking of the column in initial experiments yield the 68m Pb\textsuperscript{204} in good purity. It is planned to extend these measurements to ascertain the presence or absence of a lead activity which is formed by the decay of 14d Bi\textsuperscript{205} in the resin column. Subsequent experiments to detect the 70 kev X-ray from Pb\textsuperscript{206}, if it is formed by K-capture from Bi\textsuperscript{205}, are in progress, using a NaI(Tl) crystal with a differential pulse analyzer.

Previous workers were unable to detect K-capture X-rays from Pb\textsuperscript{205} presumably formed by the Pb\textsuperscript{204}(n,\gamma) reaction. The possibility of L-capture in Pb\textsuperscript{205} is being examined by filling counting tubes with (CH\textsubscript{3})\textsubscript{4} Pb made from lead carrier added to a thallium foil target irradiated with 2500 \muah of 15 Mev deuterons (Tl\textsuperscript{205}(d,2n)Pb\textsuperscript{205}). The quantitative small-scale synthesis of Pb(CH\textsubscript{3})\textsubscript{4} from LiCH\textsubscript{3} + PbI\textsubscript{2} + CH\textsubscript{3}I has been achieved. Purification of the tetramethyl lead product is being worked out. (R. H. Herber and D. H. Freeman)

B. BaCl\textsubscript{2} Analysis

It has been reported\textsuperscript{6} that BaCl\textsubscript{2} \cdot H\textsubscript{2}O precipitated from a mixture of 5 parts HCl and 1 part diethyl ether may contain up to 1.5 H\textsubscript{2}O molecules per Ba\textsuperscript{2+} ion, depending on exact conditions. It has also been found that if as much as 20\% water is present with the HCl-ether reagent, the precipitation is far from quantitative, frequently giving a chemical yield of 20\% or less. Since this method is useful in radiochemical analyses of mixtures of fission products where appreciable water may be present, it is desirable, if possible, to modify the procedure so that the presence of water does not lead to such poor chemical yields. Organic solvents other than ether are being investigated in varying proportions with concentrated HCl as precipitating reagents for Ba. (C. H. Donnelly, W. Poole, J. W. Winchester and C. D. Coryell)
IV. RADIOCHEMICAL STUDIES OF THE FISSION PROCESS

A. Fission Yields in the Valley Region

In connection with the identification of fission products and the determination of fission yields on the valley walls for thermal neutron U-235 fission, two half-lives and a maximum $\beta$ energy have been determined. The values obtained are given in the following table:

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Previous Half-life</th>
<th>Half-life from This Work</th>
<th>Range of $\beta$</th>
<th>$E_\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rh$^{105}$</td>
<td>36.5h</td>
<td>(+.08h 35.30h -.05h)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Rh$^{107}$</td>
<td>26m, 24m</td>
<td>21.6±0.1m (Sample 1) 21.6±0.1m (Sample 2)</td>
<td>-</td>
<td>580 mg/cm$^2$ Al 1.3 Mev</td>
</tr>
</tbody>
</table>

In each determination, the Ru parent was distilled from a mixture of fission products by fuming HClO$_4$. The Ru was then allowed to decay for a few half-lives producing Rh daughter activity, and again fumed with HClO$_4$ to remove all remaining Ru. The activity of the very pure Rh thus obtained was observed for nearly ten half-lives with an end-window proportional flow counter. All samples were mounted on solid Al cards, 445 mg/cm$^2$ in thickness, constituting infinite backscatterer.

The maximum $\beta$ energy of 21.6m Rh$^{107}$ was determined by observing decay of the sample through selected Al absorbers and plotting the activity at one specific time versus total absorber thickness in mg/cm$^2$ (including sample cover, air, and counter window). Since this was done early in the decay of the sample no appreciable amounts of activity other than the 21.6m Rh were present. The absorption curve was then analyzed by Feather analysis (relative to RaE) and only the single 1.3 Mev $\beta$ was present.

In determining the half-life of 35.30h Rh$^{105}$, determination of the slope of a plot of log (activity) vs time did not give accuracy as great as the accuracy in the counting data itself, so a method was developed to expand the scale of the graph. In this method, an approximate half-life of 35.3h was read from the decay curve. Then, starting with the activity of the first point and using this approximate half-life, a new decay curve was calculated which may not exactly coincide with the true decay curve. Finally, the differences between the calculated decay curve and
the experimental points, with their counting errors, were plotted versus time, and the slope of this plot led to the correction to be added to the approximate half-life. This method has the advantage that experimental points with different errors can be plotted and their relative weights estimated visually which can be more accurate than a method where all points are weighed the same. However, the method is best applicable only to the decay of a single nuclide. Grateful acknowledgement is made to Dr. Daniel R. J. Boyd of the Department of Chemistry for discussions leading to this method. (J. W. Winchester)

B. Decay Properties of $^{9.7m}\text{Cs}^{139}$ and $^{30m}\text{Cs}^{138}$

Work on instrumentation preparatory to continued investigation of the decay of Cs$^{139}$ is in progress. A $\beta-\gamma$ coincidence counter in conjunction with a single channel differential discriminator is being constructed. Also end-window proportional flow counters$^7$ are now in operation.

An attempt will be made to separate rubidium from cesium by solvent extraction. The extractability of CsPF$_6$ and RbPF$_6$ into organic liquids of high dielectric constant, e.g., nitromethane and nitrobenzene, will be investigated. (R. C. Fix)

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J. W. Irvine, Jr., "Applications of Radioactivity", Mid-Hudson Section, American Chemical Society, Beacon, New York, November 30, 1954.


C. D. Coryell
J. W. Irvine, Jr.
I. ENOLIZATION OF KETONES

The kinetics of racemization of D(+) - isobutyladesoxybenzoin in acetate buffer solutions as mentioned in the LNS Progress Report, November 30, 1954, has been extended to a ten to one buffer ratio (acetic acid to acetate ion). Surprisingly enough, acetic acid has proved to be a very weak catalyst. For example, with 0.030 M sodium acetate and 0.300 M acetic acid the first order rate constant is $3.20 \times 10^{-7}$ sec$^{-1}$ (at 97.9°C.), whereas with the same acetate ion concentration but a ten-fold decrease in acetic acid (0.030 M), the first order rate constant is $2.80 \times 10^{-7}$ sec$^{-1}$. Part of this small difference must be due to the ten-fold decrease in hydronium ion concentration. It is estimated that in the 1:1 buffer ratio solutions, the hydronium ion concentration is ca. $2 \times 10^{-6}$ M whereas in the 10:1 buffer solution, the hydronium ion concentration is $2 \times 10^{-5}$ M.

Some difficulty was encountered in the determination of the rate of racemization with hydroxide catalysis and this work is still in progress. The work on the catalytic effect of hydronium ion, however, has been completed and the following expression has been found:

$$k_{H_3O^+} = k_1/ (H_3O^+) = 2.48 \pm 0.11 \times 10^{-4} \text{ M}^{-1} \text{ sec}^{-1} \text{ (at 97.9}^{\circ}\text{C.})$$

D(+) - isobutyladesoxybenzoin was equilibrated with tritium by refluxing the ketone with tritiated water, hydrochloric acid, and p-dioxane. The labeled dl-ketone was isolated and purified. Reaction solutions were then made up identically to those used for racemization studies and placed in sealed bulbs. At various time intervals the bulbs were removed from the thermostat bath and the radioactivity of the water present in the sample was determined. Thus, the rate of increase of activity in the water is a measure of the rate at which the tritiated ketone enolizes. By comparing this rate to the rate of racemization (the rate at which the protonated ketone enolizes), a value for the tritium isotope effect was obtained.
The tritium isotope effect ($k_H^\text{enolization} / k_T^\text{enolization}$) has been determined for hydronium ion catalysis (with the solvent acting as the base), as shown in Table 3.1.

TABLE 3.1

<table>
<thead>
<tr>
<th>(H$_2$O)$_2$+</th>
<th>$k_H^\text{enolization}$</th>
<th>$k_T^\text{enolization}$</th>
<th>$k_H^\text{enolization} / k_T^\text{enolization}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0600</td>
<td>15.40</td>
<td>1.609</td>
<td>9.57</td>
</tr>
<tr>
<td>0.0400</td>
<td>10.10</td>
<td>1.053</td>
<td>9.59</td>
</tr>
<tr>
<td>0.0200</td>
<td>4.91</td>
<td>0.520</td>
<td>9.44</td>
</tr>
</tbody>
</table>

Average = 9.53 ± 0.06

Tritium isotope effects for acetate, hydroxide, and acetic acid catalysis are in progress as well as the determination of the ratio of the product terms (acetic acid times acetate ion) between hydrogen and tritium enolization. (E. C. Stivers)

II. THE RATE OF PROTON TRANSFER BETWEEN THE HYDROXYL GROUPS OF ALCOHOLS

The rate of hydrogen exchange between tert-butanol and triphenyl-carbinol-t had been shown to be measurable by a flow method and was found to be catalyzed strongly by benzoic acid (LNS Progress Report, November 30, 1954).

Further experiments indicated that the exchange is also base-catalyzed. Thus, on mixing toluene solutions of triphenylcarbinol-t with tert-butanol containing tri-n-butylamine (distilled from lithium aluminum hydride), and passing the mixture through a 91 cm. long flow tube, 80 percent exchange was obtained under conditions which resulted in only 40 percent exchange in the absence of the amine.

In order to find out to what extent the rate of hydrogen exchange between alcohols depends on steric hindrance by bulky substituents close to the hydroxyl groups, the hydrogen exchange of tert-butanol (0.10 M) with 2, 6-di-tert-butyl-p-cresol-O-t (0.13 M) in toluene solution was studied. In a flow tube allowing 0.17 sec. of contact, the exchange was complete; in a very short flow-tube, giving about 0.01 sec. contact time, the exchange was 70 percent complete. Thus the
phenol, in spite of its two bulky ortho tert-butyl groups, is much more reactive than triphenylcarbinol towards hydrogen exchange with tert-butanol.

For the liquid scintillation counting of tert-butanol-t at 0°, a solution of 2, 5-diphenyloxazole in toluene was used.

Experiments are being made to determine the kinetic order of the exchange with respect to the component alcohols and with respect to acid and base catalysts. (M. M. Halmann)

III. THE THERMAL DECOMPOSITION OF BENZOYL PEROXIDE

It has been found that a cyclohexane solution of benzoyl peroxide probably gives no more than 4 percent phenyl benzoate in a cage reaction.

A 0.03 M solution of tritium-labeled benzoyl peroxide containing sufficient styrene (0.15 M) to give a minimum decomposition rate was kept at 90° for 20 hours. Known amounts of non-radioactive phenyl benzoate and diphenyl were then added to the reaction mixture, thoroughly mixed, and reisolated. The diphenyl has not yet been completely purified. The phenyl benzoate was chromatographed twice over silica gel. The tritium content of this product indicated that 3.9 percent of the benzoyl peroxide has formed phenyl benzoate in a cage reaction.

It may be that the activity in the phenyl benzoate is due to an impurity. To check this, non-radioactive benzoyl peroxide will be decomposed as above. The active phenyl benzoate will be added to and reisolated from the mixture. If the activity in the phenyl benzoate is not due to impurities, the activity of this reisolated material will be decreased by only about 4 percent. If the activity is due to impurities, the activity of the reisolated phenyl benzoate will probably be lowered much more than this. The method will fail if the impurity is formed in small amount and if the whole of it remains with the phenyl benzoate during purification.

Tritium labeled benzoyl peroxide was prepared according to the scheme
Nuclear Chemistry (Organic) Group

\[
\text{C}^\text{O} \text{Cl} \quad \xrightarrow{\text{Na}_2\text{O}_2 \quad 65\%} \quad \text{C}^\text{O} \quad \text{O} \quad \text{O} \quad \text{C} \\
\text{T} \quad \text{T} \quad \text{T}
\]

(2.68 mc/MM)

The isolation and purification of phenyl benzoate from the benzoyl peroxide decomposition, after dilution with the inactive ester, was carried out by repeated chromatography over silica gel (see LNS Progress Report, November 30, 1954), and two crystallizations from petroleum ether (m.p. 69-71°, specific activity 0.061 mc/MM). (L. J. Schaad)

ADDRESSES


J. C. Sheehan
C. G. Swain
Cosmic Ray Group

I. OPERATION OF THE MULTIPLATE CHAMBER AT ECHO LAKE

New scintillation counters have been installed in the top and bottom of the cloud chamber, and the triggering system described in the LNS Progress Report of August 1954 is in operation.

The first pictures are in the process of analysis to determine whether or not the detector operates as expected. Work has also begun to determine whether or not a determination of ionization through drop counting is feasible with the new optical system. (H. S. Bridge, H. C. DeStaebler, R. A. Hewitt and W. B. Smith)

II. ANALYSIS OF CLOUD CHAMBER RECORDS

The analysis of the pictures obtained with the MIT multiplate cloud chamber at Echo Lake, Colorado, has continued. A number of new interesting examples of S-events has been found. It is becoming increasingly clear that the S-events represent decay processes of two different kinds of particles; that is, a $K_{\pi^2}$-particle (also called $\chi$-meson or $\theta^\pm$ particle) which decays into a $\pi^\pm$ meson and a $\pi^*$-meson:

$$K_{\pi^2} \to \pi^+ + \pi^*,$$

and a $K_{\mu^2}$-particle decays into a $\mu$-meson and a neutrino:

$$K_{\mu^2} \to \mu + \nu.$$

The work is being continued in an effort to determine more accurately the range of the secondary particles, and therefore the masses of the primary particles. At present it appears that the masses of $K_{\pi^2}$- and $K_{\mu^2}$-particles are close to one another and close to the $\tau$-meson, even though it seems that a small difference exists between the mass of the $K_{\mu^2}$-particle and that of the $\tau$-meson.

The investigation of possible angular correlation in the decay of $V^*$ particles is still under way, but no definite results have been obtained so far. (E. Boldt, H. C. DeStaebler, B. V. Sreekantan, G. Sandri, A. Pevsner and B. B. Rossi)
III. OPERATION OF THE LARGE MULTIPLATE CLOUD CHAMBER
AT BROOKHAVEN NATIONAL LABORATORY

No new data have been obtained during the past three months because the cosmo­tron has been shut down for this period. Machine operation is expected to com­mence in April or May and advantage has been taken of the shut-down to make various modifications in the experimental arrangement.

The preliminary runs indicate that much better beam collimation and control of the beam intensity were needed. Work has been done on both of these problems and it is hoped that the new experimental arrangements will be satisfactory.
(H. S. Bridge, H. C. DeStaebler, A. Pevsner and B. B. Rossi—MIT; in collaboration with C. Leavitt and D. Willard of Brookhaven National Laboratory)

IV. HIGH PRESSURE RECTANGULAR IONIZATION CHAMBER

The experimental work in Colorado was completed in January. Data were taken at each of three stations. Four thousand sixty-five events were recorded at the top of Mount Evans (mean barometric pressure of 46 cm Hg.) during a running time of 155.2 hours, 6000 events at Echo Lake (mean barometric pressure of 51.7 cm Hg.) during a running time of 591.5 hours, and 2724 events at Idaho Springs (mean barometric pressure of 58 cm Hg.) during a running time of 573.7 hours.

A rough analysis of the data has already been performed, so that the events have been separated according to pulse height distribution and discharges in the top and bottom trays. Preliminary calculations indicate that the mean free path in the atmosphere for nucleons with an energy of 200 Bev or greater is approximately 110 g/cm² (corrected for Gross transformation). More detailed analysis and calculations are now in progress. (Leonilda Altman)

V. AIR SHOWER MONITOR

The monitor in Colorado continues to operate with little trouble. C. S. Robertson has begun the analysis of the data, which should be comparable with that of Daudin and Daudin.¹
Ricardo Palmeira of the Brazilian Center of Physics and the Chacaltaya Laboratory has joined the group and will complete the already-started work on the equipment for a similar monitor to go to Chacaltaya (Bolivia). (R. D'Arcy, R. A. Hewitt, R. Palmeira, C. Robertson and R. W. Williams)

VI. HIGH ENERGY CROSS SECTIONS

Construction of equipment for a new measurement of the collision cross section of \(10^{11}\) ev nucleons is under way.

The nuclear model and nucleon-nucleon cross section described in LNS Progress Report lead to a collision mean free path in air of 79 g cm\(^{-2}\). Interpretation of the difference between this and the observed absorption mean free path (usually taken as 125 g cm\(^{-2}\); recent results indicate it may be smaller -- see Section IV of this report) is being re-examined to see if it is consistent with the large value of the nucleon-nucleon cross section. (A. Brenner and R. W. Williams)

VII. LARGE AIR SHOWER EXPERIMENT

Twenty large scintillation counters of the type previously described have been moved to the Harvard Observatory at Harvard, Massachusetts and located in a symmetrical array over an area of fifty acres. The electronic recording equipment is assembled in the Quonset hut and the cables connecting the counters to the hut are laid. Final testing of the equipment is now under way.

In order to measure the relative sensitivity of the photomultipliers which are to be used in the twenty-counter array, we have built a cylindrical counter 1 m\(^2\) in area and 2 m tall with sockets for 9 photomultipliers in the top. One of these photomultipliers is run at low voltage, so that we are confident that no space-charge limitation occurs in this photomultiplier for large air shower pulses. The other photomultipliers are run under regular operating conditions and their responses to air shower pulses are simultaneously recorded. In this way we can determine the relative sensitivity of the photomultipliers over a wide range of pulse sizes.

An electronic integrator has been constructed for use in calibrating the counters with \(\mu\)-mesons. This device measures the average pulse height produced in a scin-
Cosmic Ray Group

tillation detector by $\mu$-mesons selected by a counter telescope. We have also completed circuits for monitoring the rate of single large pulses from each counter. These rates will provide a check on variations in the relative sensitivity of the counters.

We are investigating the possibilities of analyzing the air shower data with an electronic computer. An empirical formula (with four adjustable parameters corresponding to the size, shape and core location of a shower) is to be fitted by the method of least squares to the pulse size data from the counters. Several methods of performing the computation are being tested for rapidity and uniqueness of convergence. The effect of the orientation of the shower axis as determined by timing measurements is to be included in the computation. After preliminary tests have been completed, a machine analysis of the data obtained with eight density detectors and four timing counters on the roof of Building 6 of MIT will be attempted. (B. B. Rossi, G. W. Clark, J. A. Earl, J. Linsley, M. Oda and F. Scherb)

VIII. TIMING MEASUREMENTS

We are now analyzing the data obtained with improved timing equipment on the distribution of arrival times of air shower particles. More accurate information will be obtained on the shape of the electron disc, the longitudinal distribution of electrons in the disc, and the zenith angle distribution of the shower axes. (G. W. Clark)

IX. DIFFUSION EQUATION OF NUCLEONIC COMPONENT

The downward diffusion of the nucleonic component of high-energy cosmic rays in the atmosphere is being investigated from the theoretical point of view. The equation describing this process is of the following type:

$$\frac{\partial N(E, x)}{\partial x} = -\frac{1}{\lambda} N(E, x) + \frac{1}{\lambda} \int K(E, E') N(E', x) + Q(E, x) \quad (1)$$

where $N(E, x)$ is the differential vertical intensity of nucleons with energies between $E$ and $E + dE$ at the atmospheric depth $x$, $\lambda$ is the collision mean free path, $K(E, E')$ is the differential multiplicity of nucleons with energies between $E$ and $E + dE$ produced by a nucleon of energy $E'$ and $Q(E, x)$ is the source function of
nucleons produced by other components (mostly \( \pi \)-mesons). At present, a new method of solving the above integro-differential equation is being tested. Preliminary results indicate that one can find a general analytic solution of Eq. (1) provided one introduces reasonable approximations in the kernel function, \( K(E, E') \). It turned out, e.g., that a kernel incorporating the basic features of Fermi's model of multiple production of mesons and (possibly) anti-nucleons can be represented in sufficiently simple form as to allow a general solution of Eq. (1). The mathematical expressions thus obtained for \( N(E, x) \) are being evaluated numerically for the purpose of comparison with experimental data. (S. Olbert)

X. INVESTIGATION OF IONIZATION MEASUREMENTS IN MULTIPLATE CLOUD CHAMBERS AND STUDY OF RANGE-ENERGY RELATION AS APPLIED TO MASS DETERMINATIONS

In order to determine more accurately the masses of some of the new particles, the status of the range-energy relation has been examined critically, and a method has been developed for utilizing the observed change in the ionization of a particle traversing a multiplate cloud chamber to give a more accurate value for that particle's range.

Nomograms have been developed to give the residual range of a particle of known mass from a measurement of the amount of material the particle has passed through in traversing a plate in the cloud chamber, and from the ratio of its ionization on each side of that plate. The result is almost independent of the type of material in the plate or in the gas, and does not depend very much on the mass of the particle. Using tracks having a residual range known to about \( \pm 1 \text{ g/cm}^2 \), it was found that visual estimates of the ionization ratio gave the residual range with an error of about \( \pm 2 \text{ g/cm}^2 \). To improve the accuracy, a photometric measuring system has been put in operation. For films with little background fog and on which calibration tracks can be found, ionization measurements can be made which are apparently limited in accuracy mainly by the statistical fluctuations in the ionization.

Mass values for unstable particles can sometimes be determined from the observed ranges of the charged secondary particles if the decay scheme is known. The accuracy of the range-energy relation is an important factor in determining the error in the mass values obtained in this way.
Cosmic Ray Group

By applying various corrections to the data (principally those for the nonparticipation of electrons in the L and higher shells), it has been possible to show that all of the accurate experiments on range, energy loss, and relative stopping power in the proton energy region from 18 to 100 Mev yield mean excitation potentials in good agreement with each other. Hence the principal remaining evidence for the mean excitation potential's being velocity-dependent is the Berkeley range measurement at 300 Mev. The mass of some particles (e.g., $K_{\mu 2}$) depends markedly upon whether or not this evidence is accepted. (D. O. Caldwell)

REFERENCES


ADDRESSES


B. Rossi, "Review of Results on S-Particles", given at Rochester Conference, January 31, 1954.

PUBLICATIONS


I. PHENOMENOLOGICAL THEORY OF THE PHOTODISINTEGRATION OF THE DEUTERON

Analysis of the data of the University of Illinois and California Institute of Technology (unpublished) on the photodisintegration of the deuteron has been made on the assumption that, at energies greater than ~50 Mev, the main processes arise from meson production and re-absorption. Two processes have been considered. In the first, the meson is produced, by electric dipole absorption, in an S-state around one of the nucleons. This meson is then absorbed by the other nucleon through the 3.3 resonance. This leads to a final state which is a mixture of $^3P_0$ and $^3P_1$ with the angular distribution $A + \cos^2\Theta$, where $A \sim 4$.

The other process involves magnetic dipole or electric quadrupole photon absorption by one of the nucleons into the 3.3 state, and the subsequent disintegration into a $^1D_2$ nucleon state, with an angular distribution of $5.3 \cos^2\Theta$. To account for the low energy data, additional, conventional, spin-independent electric dipole absorption is required.

These three processes can be used to fit the observations with reasonable values of the cross sections and of their energy dependence. The asymmetry in the angular distribution is assumed to arise from a retardation effect in the electric dipole absorption processes. An interesting point is that if this interpretation should prove to be reasonable it would enable an unambiguous evaluation of the strength of the electric quadrupole absorption in the 3.3 state, since this would give rise to an appreciable $\cos^4\Theta$ term in the angular distribution. (B. T. Feld)

II. PHOTONUCLEAR RESEARCH

A. Photon Scattering

1. Low Energies

Analysis of the photon scattering data between 0.6 and 3.25 Mev obtained with the Building 28 vertical Van de Graaff generator has now been completed. The mea-
Elementary Particle Scattering Group

measured elastic (or near-elastic) differential scattering cross section per atom at 90° for the four elements studied are shown in Fig. 5.1, along with the measured points of other experimenters in this energy range. The cross sections are plotted absolutely, normalized by actual measurements of the bremsstrahlung flux from the Van de Graaff. For comparison, the inelastic Compton scattering cross sections per atom are of the order of a few times $10^{-25}$ cm²/steradian.

The rapidly decreasing scattering component below 2 Mev is interpreted as Rayleigh scattering by bound electrons, although the measured magnitude is appreciably greater than that calculated from Bethe's form factor (higher by ~5 percent in Pb, ~8x in Cu at 1 Mev). Since the form factor result is only approximate and the exact numerical values for mercury obtained by the Birmingham group are higher than the form factor of expression, it was assumed for analysis that the measured cross section at low energies represents the true Rayleigh scattering. This measured Rayleigh cross section was combined with the calculated nuclear Thomson cross section and the result subtracted from the measured curves, the difference presumably representing scattering due to nuclear excitation. (Delbruck scattering might also contribute - its magnitude at large angles has not been calculated.)

Using the above subtraction procedure, the nuclear scattering at 3 Mev is found to be ~15 µbarns/steradian at 90° in In, 12 in Cd, and 5 in Cu, with uncertainties of the order of a factor of two (the nuclear scattering in Pb is too small compared to the Rayleigh for the subtraction to be reliable.) These values are in reasonable agreement with measured isomer photo-excitation cross sections, although uncertainties in comparison prevent the drawing of detailed conclusions, and they are also consistent with the scattering measurements at higher energies made at the National Bureau of Standards by Fuller and Hayward (whose lowest-energy points are shown in Fig. 5.1). The various theories of photonuclear reactions are not sufficiently developed to make definite predictions.

2. Higher Energies

The investigation of the scattering and absorption of the 4-17 Mev X-rays produced by the linear accelerator is being continued. The photon detector, a scintillation pair spectrometer, has been completed. Preliminary testing of the
Figure 5.1

Differential Elastic Scattering Cross Sections at 90°
Elementary Particle Scattering Group

Rockefeller generator using the \((p, \gamma)\) reaction in boron indicates that the counter energy resolution is of the order of 10 percent.

B. **Bremsstrahlung Spectrum**

An experiment is in progress to measure the thin target bremsstrahlung spectrum from the analyzed beam of the linear accelerator. The energy spectrum of photons produced by photodisintegration of deuterium will be obtained by means of an anthracene crystal scintillation counter, and the bremsstrahlung spectrum will then be found by using the known cross section for the deuteron disintegration. The polarization of the bremsstrahlung beam will also be investigated.

C. **Photoneutron and Photoproton Production**

Machine difficulties have prevented the procurement of final data for the total neutron yield experiment previously described. This work is continuing, as is the search for a suitable neutron spectrometer for use in measuring photoneutron energy spectra.

The photoproton experiment is ready to begin, as soon as the total neutron yield work is completed. Both scintillation counters and photographic plates will be used to detect the protons, and it is hoped that both yields and energy spectra can be obtained.

D. **Ba\(^{137}\) Isomer Excitation**

An experiment has been started at Building 28 to measure the excitation curve for photoproduction of the isomeric state of Ba\(^{137}\) by thick target bremsstrahlung. The isomeric activity produced will be counted with a scintillation counter (2.3 minute half-life). A search will be made for breaks in the slope of excitation curve, corresponding to energy levels in the excited Ba\(^{137}\) nucleus.

E. **Bremsstrahlung Tables**

Numerical values of the Bethe-Heitler bremsstrahlung cross section have been computed and tabulated for electron energies from 1.50 to 15.50 Mev and photon energies from 1.25 to 15.25 Mev, both in 0.25 Mev intervals. The cross sections were calculated for a lead radiator, but can be used for any other heavy element by appropriate scaling. These tables have been lithographed and are
available on request to the Laboratory for Nuclear Science headquarters. (W. Bertozzi, J. L. Burkhardt, P. T. Demos, T. H. Dupree, F. Paolini, C. P. Sargent, W. J. Sawtelle and J. E. Snyder)

III. PHASE SHIFT ANALYSIS OF THE $^{16}\text{O}^{16}(p,p)^{16}\text{O}$ EXPERIMENT

The coding of the problem described briefly in the LNS Progress Report of August 31, 1954 has been completed by the Joint Computing Group and checked out on the Whirlwind computer. The absolute cross-section data have been re-examined for possible sources of error which may have been overlooked up to now and currently the data at the eight scattering angles are being replotted as a function of proton bombarding energy. The energy range from .500 Mev to 2.6 Mev will be the first to be examined. At about ten energies within this interval, attempts will be made to fit the angular distributions obtained from the cross-section data with various combinations of phase shifts selected from among the following: $\delta_0, \delta_1^{\pm}, \delta_2^{\pm}, \delta_3^{\pm}$. For any given angular distribution attempts will be made to establish the uniqueness of fit and to establish the uncertainty in any given set of phase angles found to fit a given distribution which are compatible with the experimental uncertainties in the data. (Elizabeth Campbell and F. J. Eppling)

J. R. Zacharias
A new technique has been developed for the detection of the de-excitation gamma rays following the inelastic scattering of neutrons, which is suitable for observing gamma rays in the energy region 100 to 350 kev. Low lying energy levels have been observed in eight elements ranging from manganese to gold. The excitation functions and cross sections for the production of these gamma rays have been measured using neutrons between 100 and 1800 kev.

A thin NaI(Tl) crystal is used as a gamma ray detector. The crystal is less than 1 mm thick, eliminating to a great extent the background problems encountered in low-energy gamma-ray measurements made in the presence of a high neutron flux. The scattering sample and crystal are shielded with 1/8 inch of lead, and placed directly in the neutron beam. Neutrons are produced by the $^3$He $(p, n)$ reaction, using protons from the Rockefeller generator. Neutron resolution is about 35 kev.

Excitation curves for the elements studied are shown in Figs. 6.1-6.8. The absolute values of cross section given represent cross sections for the excitation of the gamma rays observed. The excitation of other levels in the nucleus contributes in several ways to the observed gamma rays.

The yield curve for the first excited state in iodine (Fig. 6.2) shows clearly the effect of competition from higher levels. Beside the 60-kev level in this element there are three known higher levels, at 214, 420, and 630 kev. Cascade radiation between these levels and the first level has not been observed. As the neutron energy is increased the higher states are excited in competition with the first excited state, causing a decrease in the excitation of the 60-kev level, as shown by decrease in the yield of the 60 kev gamma ray for high neutron energies.

The yield curve for the first excited state in tantalum (Fig. 6.7) shows the effect of cascade transitions to this state from higher excited states. Here the yield is the sum of the de-excitation radiation from the 137-kev first level and the radiation following cascade transitions from higher levels. Actually the situation
Neutron Physics Group

Figure 6.1

Neutron Inelastic Cross Section for Manganese

Figure 6.2

Neutron Inelastic Cross Section for Iodine

Figure 6.3

Neutron Inelastic Cross Section for Gold

Figure 6.4

Neutron Inelastic Cross Section for Platinum
Figure 6.5
Neutron Inelastic Cross Section for Rhenium

Figure 6.6
Neutron Inelastic Cross Section for Wolfram

Figure 6.7
Neutron Inelastic Cross Section for Tantalum

Figure 6.8
Neutron Inelastic Cross Section for Hafnium
Neutron Physics Group

is even more complicated. The second excited state in tantalum is at about 300 kev. Thus the cascade transition from this level to the 137 level has an energy of 163 kev, and cannot be resolved by the spectrometer from the 137-kev radiation. The gamma radiation observed then represents the sum of the excitation of the 137-kev level, and twice the excitation of the 300-kev level, assuming that this decays entirely by cascade. The continued rise of the yield curve above threshold energies is therefore probably caused by cascade transitions.

From the above discussion it is clear that a complete knowledge of the decay schemes of the elements under observation is essential for an exact evaluation of the yield curves in terms of cross section. In the case of hafnium (Fig. 6.8) which has five stable isotopes, the observed gamma radiation probably contains contributions from cascade and direct-to-ground transitions in all isotopes, since the energies of many of these transitions overlap. Here, the use of separated isotopes would help to clear the interpretation.

For five of the elements investigated theoretical yield curves were calculated from the theory of Hauser and Feshbach. These are shown dotted in the figures. In the calculation penetrabilities $T_\ell$ calculated using the black nuclear model were used. In these calculations, all orbital angular momenta up to $\ell = 3$ were considered. In every case the assumption was made that a single level was being excited, and that the statistical model is valid for the compound nucleus, but not for the residual nucleus. A summary of results, and comparison with theory is presented in Table 6.1.

One of the reasons for studying the elements that were chosen for this investigation was to determine the sensitivity of the inelastic neutron scattering cross section to the rotational level hypothesis of Bohr and Mottelson. Table 6.1 shows certain general trends as one goes farther away from the closed-shell nucleus Pb$^{208}$ toward the region of high nuclear deformation where the rotational model is most applicable. Agreement of the absolute values of cross section with those predicted by theory gets better, while shape agreement gets worse. Absolute values of cross section increase. To some extent these trends may be explained by the increasing density of low-lying levels, and to the increasing probability of cascade transitions. This does not explain, however, the slow rise of the yield curves near threshold for the elements wolfram, tantalum, and hafnium. We conclude that for those elements known to have rotational level schemes the excitation process is modified from that predicted by Hauser-Feshbach theory.
TABLE 6.1

Summary of Results

<table>
<thead>
<tr>
<th>Element</th>
<th>A</th>
<th>Eγ (kev)</th>
<th>Fit of Shape to Hauser-Feshbach</th>
<th>$\sigma_{\text{exper}}/\sigma_{\text{H-F}}$</th>
<th>$\sigma_{\text{in}}$ (barns) $E_n = 1.5$ Mev</th>
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<tbody>
<tr>
<td>79Au</td>
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<td>Good</td>
<td>0.27</td>
<td>0.36</td>
</tr>
<tr>
<td>78Pt</td>
<td>even</td>
<td>330</td>
<td>Fair</td>
<td>0.36</td>
<td>0.62</td>
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<tr>
<td>75Re</td>
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<tr>
<td>74W</td>
<td>mostly even</td>
<td>115</td>
<td>Poor</td>
<td>-</td>
<td>- 0.95</td>
</tr>
<tr>
<td>73Ta</td>
<td>odd</td>
<td>137</td>
<td>Poor</td>
<td>1.25</td>
<td>1.50</td>
</tr>
<tr>
<td>72Hf</td>
<td>odd + even</td>
<td>230</td>
<td>Poor</td>
<td>-</td>
<td>2.4</td>
</tr>
</tbody>
</table>

(Just B. Guernsey)

II. ELECTRIC EXCITATION OF CADMIUM, PALLADIUM AND MOLYBDENUM

Energy levels in enriched isotopes of cadmium, palladium and in natural molybdenum have been studied by electric excitation with protons. The observed $\gamma$-rays together with the electric excitation cross sections and the reduced electric quadrupole transition probabilities are shown in Table 6. II.

The pulse height spectrum produced by $\gamma$-rays from powdered Pd and enriched Pd$^{106}$ targets during bombardment with 2.46 Mev protons is shown in Fig. 6. 9. Peaks corresponding to $\gamma$-rays having energies of 365, 425 and 500 kev from natural Pd and 220, 430 and 500 kev from enriched Pd$^{105}$ are observed. The 270 kev line comes from electric excitation of Pd$^{105}$ and probably corresponds to the known level at 282 kev in this isotope observed$^5$ in the K-capture decay of Ag$^{106}$. The line at 500 kev is due to electric excitation of 27.1 percent abundant Pd$^{106}$ and can also be identified with a known level in this isotope at 500 kev which is observed$^5$ in the K-capture decay of Ag$^{106}$. The 500 kev $\gamma$-ray is also observed from the enriched Pd$^{106}$ sample with 0.6 the intensity of the $\gamma$-ray from natural Pd since the enriched sample contains 16.04 percent Pd$^{106}$. The 425 and 430 kev $\gamma$-rays are more difficult to assign to the proper isotopes. The most
The pulse height spectra obtained when natural Pd and enriched Pd$^{105}$ targets are bombarded with 2.46-Mev protons are shown in this figure. The principal isotopes present in the enriched Pd$^{105}$ target are Pd$^{105}$, 78.19 percent, and Pd$^{106}$, 16.04 percent. The peak at 425 kev from the natural Pd target is a composite line caused by $\gamma$-rays of nearly equal energies in Pd$^{105}$, and Pd$^{108}$. A 0.04 inch Cd absorber was used to attenuate the palladium X-rays. The broad low-energy pulse distribution is due to proton bremsstrahlung.

Figure 6.9
TABLE 6. II

Summary of Results

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Measured Excitation Energy (kev)</th>
<th>Proton Energy (Mev)</th>
<th>$\sigma$ (Thin) Millibarns</th>
<th>$\sigma$ (Thick) Millibarns</th>
<th>$I_0 \rightarrow I$</th>
<th>$\Delta E$</th>
</tr>
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<tbody>
<tr>
<td>V 51</td>
<td>325</td>
<td>1.50</td>
<td>-</td>
<td>.47</td>
<td>7/2→</td>
<td>?</td>
</tr>
<tr>
<td>Cr 53</td>
<td>155(?)</td>
<td>1.50</td>
<td>-</td>
<td>.12</td>
<td>7/2→</td>
<td>? .15</td>
</tr>
<tr>
<td>Mo 55</td>
<td>131</td>
<td>.94</td>
<td>.29±. 11</td>
<td>-</td>
<td>5/2→</td>
<td>? .87</td>
</tr>
<tr>
<td>Mo 95</td>
<td>199</td>
<td>2.46</td>
<td>-</td>
<td>.043</td>
<td>5/2→</td>
<td>? .57</td>
</tr>
<tr>
<td>Pd 105</td>
<td>270</td>
<td>2.46</td>
<td>-</td>
<td>.02a</td>
<td>5/2→</td>
<td>? .084</td>
</tr>
<tr>
<td></td>
<td>430</td>
<td>2.75</td>
<td>-</td>
<td>.10a</td>
<td>5/2→</td>
<td>? .57</td>
</tr>
<tr>
<td>Pd 106</td>
<td>500</td>
<td>2.75</td>
<td>-</td>
<td>.080</td>
<td>0 →</td>
<td>2 1.8    49</td>
</tr>
<tr>
<td>Pd 108</td>
<td>425</td>
<td>2.75</td>
<td>-</td>
<td>.15b</td>
<td>0 →</td>
<td>2 2.1    58</td>
</tr>
<tr>
<td>Pd 110</td>
<td>365</td>
<td>2.75</td>
<td>-</td>
<td>.10</td>
<td>0 →</td>
<td>2 2.6    67</td>
</tr>
<tr>
<td>Ag 107, 109</td>
<td>315</td>
<td>2.75</td>
<td>.27b</td>
<td>.24b</td>
<td>1/2→</td>
<td>3/2 .79   18</td>
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<tr>
<td></td>
<td>418</td>
<td>2.75</td>
<td>.20b</td>
<td>.19b</td>
<td>1/2→</td>
<td>5/2 .86   27</td>
</tr>
<tr>
<td>Cd 111</td>
<td>330</td>
<td>2.75</td>
<td>-</td>
<td>.010</td>
<td>1/2→</td>
<td>3/2 .27   -</td>
</tr>
<tr>
<td>Cd 112</td>
<td>610</td>
<td>2.75</td>
<td>-</td>
<td>.028</td>
<td>0 →</td>
<td>2 1.3    43</td>
</tr>
<tr>
<td>Cd 113</td>
<td>290</td>
<td>2.75</td>
<td>-</td>
<td>.020</td>
<td>1/2→</td>
<td>3/2 .58   -</td>
</tr>
<tr>
<td>Cd 114</td>
<td>545</td>
<td>2.75</td>
<td>-</td>
<td>.050</td>
<td>0 →</td>
<td>2 1.4    48</td>
</tr>
<tr>
<td>In 115</td>
<td>500</td>
<td>3.00</td>
<td>-</td>
<td>.010</td>
<td>9/2→</td>
<td>? .58    -</td>
</tr>
<tr>
<td>I 127</td>
<td>212</td>
<td>2.00</td>
<td>-</td>
<td>.010</td>
<td>5/2→</td>
<td>? -       -</td>
</tr>
</tbody>
</table>

a = Enriched isotope target
b = Composite line

The experimental results are summarized in this table. Column 3 lists the proton energy at which the cross-section measurement for each level was made. In column 6, the spin change of the transition is shown for those cases in which it is known or can be reasonably inferred from the collective model. Column 7 shows the value of B(E2) for the excitation of the levels obtained from the measured cross sections shown in columns 4 and 5 and from the measured energies shown in column 2.
probable interpretation is that the 430 kev $\gamma$-ray seen in the enriched $\text{Pd}^{105}$ comes from electric excitation of a level in $\text{Pd}^{105}$ at this energy while the 425 kev $\gamma$-ray seen in natural Pd is a composite line caused partially by excitation of the 430 kev level in 22.6 percent abundant $\text{Pd}^{105}$ and of a known level at about this energy in 26.7 percent abundant $\text{Pd}^{108}$ which is observed in the K-capture decay of $\text{Ag}^{108}$. The 365 kev line in natural Pd is due to electric excitation of a level in 13.5 percent abundant $\text{Pd}^{110}$ which has been identified by Temmer and Heydenburg.

The pulse height spectra produced when targets of CdO, enriched Cd$^{111}$O, and enriched Cd$^{113}$O are bombarded with 2.46 Mev protons are shown in Fig. 6.10. The $\gamma$-ray at 330 kev is due to electric excitation of a level in Cd$^{111}$ at about this energy which is observed in the $\beta^-$ decay of $\text{Ag}^{111}$. The line at 610 kev which appears in the enriched sample and also in the natural target is due to electric excitation of a level in Cd$^{112}$. The 290 kev peak is caused by electric excitation of a level in Cd$^{113}$ and the line at 545 kev which appears in the enriched Cd$^{113}$O sample and also in natural CdO is due to electric excitation of Cd$^{114}$. This $\gamma$-ray corresponds to a known level in Cd$^{114}$ at 548 kev which is observed in the K-capture decay of In$^{114}$. It is improbable that the 420 and 500 kev $\gamma$-rays observed from all three targets arise from electric excitation of any Cd isotope. They are probably the result of bombardment of oxygen with protons since they appear with roughly equal intensities from all targets.

These $\gamma$-rays (199, 440 and 525 kev) are observed when a powdered molybdenum metal target is bombarded with 2.46 Mev protons. The $\gamma$-ray at 199 kev has been observed previously by Temmer and Heydenburg and is probably due to electric excitation of a level at about this energy in Mo$^{95}$ which has also been observed in the positron decay of Te$^{85}$. The origin of the 440 and 525 kev $\gamma$-rays is not clear. It is probable that the 440 kev line is due to a sodium impurity in the target. The 525 kev line has been observed by Temmer and Heydenburg using 6 Mev $\alpha$-particles. The isotopic assignment of this line is in doubt.

Table 6.11 summarizes the experimental results for these and other nuclei which have been investigated recently. (H. Mark and G. Paulissen)
The pulse height spectra obtained when natural CdO, enriched Cd\textsuperscript{110}O, and enriched Cd\textsuperscript{13}O targets are bombarded with 2.46-Mev protons are shown in this figure. The principal isotopes present in the Cd\textsuperscript{110}O target are Cd\textsuperscript{111}, 64.5 percent and Cd\textsuperscript{113}, 26.9 percent and the principal isotopes present in the Cd\textsuperscript{13} target are Cd\textsuperscript{113}, 54.1 percent and Cd\textsuperscript{114}, 37.9 percent. The peaks at 420 and 500 kev that appear in all three spectra are probably the result of some process following the bombardment of oxygen with protons. The broad low-energy pulse distribution is due to proton bremsstrahlung.
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ADDRESSES


I. INTRODUCTION

During the past three months, we have continued our general program of measurements on the excited states of nuclei. The present status of those parts of the program which have been most actively pursued during this period is outlined in subsequent sections of this report.

Most of this research has been carried out with the new broad-range magnetic spectrograph. This spectrograph has been further improved and is now an exceptionally effective instrument. Scintillation counters for heavy particles have been installed in the target chamber and behind a slit in the focal plane. The first enables continuous monitoring of the beam and target during bombardment, and the second permits the use of the instrument as a high-resolution spectrometer. Additional measurements have been made on the alignment of the spectrograph with respect to the beam line, as a function of the angular position, and three sensitive dial indicators have been mounted on the magnet yoke to give a continuous and precise check on the magnet position. A low-energy background which made inelastic scattering measurements difficult, particularly at small angles, has been identified as arising from scattering from the energy defining slit at the exit of the deflecting magnet. A precision defining slit and collimator is now being made in the shop and, when installed, should greatly reduce this background of scattered particles.

II. ANGULAR DISTRIBUTION STUDIES OF \( ^{14}\text{Ni} (d, p)^{15}\text{Ni} \)

After the discovery\(^1\) that the first and second excited states of \( ^{15}\text{Ni} \) have a separation of only 29 kev, there has been continuing discussion regarding the nature of these states, which lie approximately 5.3 Mev above the ground state. In recent years, a number of investigations of the angular distributions of the proton groups from the \( ^{14}\text{Ni} (d, p) \) reaction have been made, but the resolution in these experiments has not been sufficient to separate the two groups corresponding to these states and thus to enable a clear-cut analysis of the result.
ONR Generator Group

Recently we have completed a study of the excited states of N\textsuperscript{15} from its ground state up to the neutron binding energy\textsuperscript{2}. This work was carried out with the 180-degree spectrograph. During this past quarter, angular-distribution measurements on N\textsuperscript{14}(d, p)N\textsuperscript{15} have been carried out with the new broad-range spectrograph.

A series of exposures has been made at a deuteron bombarding energy of 7 Mev and at angles between 5 and 55 degrees in 5-degree steps and between 60 and 110 degrees in 10-degree steps. At each angle, exposures of 1000 \(\mu\)c and 100 \(\mu\)c were made to permit track counting of both weak and strong groups. A thin film of nylon, supported by an evaporated layer of gold, served as target material. To check for any changes in target composition and particularly for the evaporation of nitrogen, an exposure at 50 degrees was repeated after every third angle. A total of seven normalizing exposures at 50 degrees was made with no appreciable change noted in nitrogen content after 15,000 \(\mu\)c of bombardment on one target and 8000 \(\mu\)c bombardment on a second target. However, a 15 percent difference in nitrogen content was observed between target No. 1 and target No. 2.

With the wide range in the energy spectrum provided by the three 10-inch nuclear plates in the new spectrograph, it was possible to record simultaneously during each exposure all the proton groups from the N\textsuperscript{14}(d, p)N\textsuperscript{15} reaction corresponding to the excitation region in N\textsuperscript{15} between about 5.3 and 11 Mev. Over this range, there are sixteen known groups from the N\textsuperscript{14}(d, p)N\textsuperscript{15} reaction, in addition to eleven proton groups from carbon and oxygen\textsuperscript{2}.

The plate counting on only the first eight groups from N\textsuperscript{14}(d, p)N\textsuperscript{15} has thus far been completed. All eight groups are clearly resolved at all angles except for the group corresponding to the 7.575 level in N\textsuperscript{15} which runs into some interference at small angles with a group from the O\textsuperscript{16}(d, p)O\textsuperscript{17} reaction (0.875-Mev level).

A preliminary analysis of the angular-distribution curves for these eight N\textsuperscript{15} groups show the following characteristics:

a) **Levels at 5.276 and 5.305 Mev in N\textsuperscript{15}**. Both groups are comparatively weak and show distinctively different angular distributions. The 5.276-Mev group shows a definite peak at about 40 degrees in the laboratory system, whereas the 5.305-Mev group appears isotropic between angles of 10 and 110 degrees. Previous measurements on this pair of groups have been conflicting. Gibson and Thomas\textsuperscript{3} using 8-Mev deuterons observed no rise in the cross section at small
angles and concluded no stripping process takes place to any great extent, and therefore the two levels have high spin. Recently, Eby\textsuperscript{4} with 11.9-Mev deuterons reported observing a distinct rise in the cross section at small angles, thus indicating Butler-type stripping. Neither observer, however, was able to resolve the two groups.

b) Level at 6.328 Mev in $N^{15}$. The present data for this group show a peak at about 20 degrees in the laboratory system and therefore probably indicate a momentum transfer of $\ell_n = 1$. The results of Gibson and Thomas\textsuperscript{3} were not conclusive for this group, although a slight rise at small angles is suggested from their curves.

c) Levels at 7.164, 7.309, and 7.575 Mev in $N^{15}$. The measurements of Gibson and Thomas\textsuperscript{3} probably included the cross sections of all three of these groups. They were unable to resolve the groups corresponding to the 7.164- and 7.309-Mev levels and had no knowledge of the existence of the 7.575 level at the time of their measurements. Their results showed a sharp rise at small angles, and they concluded at least one level has $\ell_n = 0$. Our present tentative curves show that each group has a distinctly different angular distribution. The 7.164 group has a peak at about 40 degrees; the 7.309 group has a peak at about 40 degrees in addition to a sharp rise at small angles; while the 7.575 group exhibits only a gradual rise toward small angles.

d) Levels at 8.315 and 8.571 in $N^{15}$. Gibson and Thomas\textsuperscript{3} reported only on the 8.315 group. They observed a sharp rise near small angles and concluded an angular momentum transfer $\ell_n = 0$. Either they did not observe the group corresponding to the 8.571-Mev level, or it was not sufficiently resolved from the 8.315 group.

Our curve for the 8.315 group is similar to that for the 7.309 group with a peak at about 40 degrees and a sharp rise near forward angles. The curve for the 8.571 group shows a gradual rise toward small angles much like the curve for the 7.575 group.

The data for the eight groups above are at present being studied in an attempt to fit the experimental curves to Butler-type stripping curves. The plate counting for the remaining eight groups from the $N^{14}(d,p)N^{15}$ reaction is continuing.

(A. Sperduto and R. Sharp)
III. ENERGY LEVELS OF Be⁹

Further work has been undertaken with the aim of corroborating the level scheme reported for Be⁹ from the Li⁷(He³, p)Be⁹ reaction. Freshly prepared targets of beryllium evaporated on formvar were bombarded with 6.00-Mev protons, and the charged-particle spectrum recorded, the spectrograph being oriented at 50 degrees to the incident beam. Unfortunately, the instrumental scattering under these conditions appeared to mask the inelastic protons from the target in the region of interest. It is hoped that the new slits mentioned previously in this report will relieve this difficulty. Meanwhile, exposures are planned at 90 degrees, where the instrumental scattering appears to be at a minimum. (C. K. Bockelman)

IV. ENERGY LEVELS OF Na²²

The Na²² nucleus is one of the interesting series of odd-odd nuclei expected to have low-lying states of isobaric spin 1. The next nucleus in this series, Al²⁶, has recently been shown to have the lowest T = 1 state very near the expected position. In this case, the state was observed by a gamma-ray, where the assignments of gamma-rays were based on knowledge of the T = 0 states as seen with the Si²⁸(d, α)Al²⁶ reaction. In the case of Na²², it was hoped that the lowest T = 1 state might be observed with a (p, α) reaction. A comparison of the states observed in this way with those found with a (d, α) reaction would give useful information on the isobaric spin-selection rule.

Little is known about the level scheme of Na²². The T = 0 levels may be conveniently investigated using the exothermic reaction Mg²⁴(d, α)Na²². A target made by evaporating a thin layer of magnesium metal onto a formvar backing was bombarded with deuterons of energies 5, 6, and 7 Mev. The alpha-particles emitted at 90 degrees were observed with the broad-range spectrograph. Nine groups of alpha-particles have been observed which can be ascribed to this reaction. The observed change of alpha energy with a change in bombarding energy plus target analysis by means of elastic scattering allowed the assignment to be limited to the Mg isotopes. Bombardment of a target enriched in Mg²⁵ eliminated this possibility and knowledge of the levels of Na²⁴ plus intensity considerations eliminated Mg²⁶.
The ground state Q-value is 1.954 ± 0.020 Mev. Preliminary values for excitation energies of some of the observed states are 0.59, 0.89, 1.53, 2.57, 2.98 and 3.06. A wide group is observed that may arise from two closely spaced levels at about 1.9 Mev. There is some indication of a level at 2.22 Mev. Further work with thinner targets and longer bombardments is in progress.

The ground state and first two excited states listed above have been observed with the Mg$^{25}$(p, α)Na$^{22}$ reaction. A target of magnesium acetate enriched in Mg$^{25}$ was bombarded with 7.5-Mev protons and the alphas observed at 60 degrees. The yield is very low; therefore longer bombardments are needed. (C. P. Browne and W. C. Cobb)

V. EXCITED STATES OF Al$^{28}$

The study of the proton groups from Al$^{27}$(d, p)Al$^{28}$ has continued using the broad-range spectrograph. Plates exposed at 6- and 7-Mev bombarding energy at 90 degrees have been analyzed in detail by M. Mazari at the National University of Mexico, and additional exposures at 10 and 35 degrees, using 7-Mev deuterons, have been taken and the plates analyzed as an additional check on the correct assignments of the proton groups.

Al$^{28}$ has by far the most complicated spectrum of bound states of any nucleus thus far investigated in detail. Previous work in this laboratory by Enge et al$^{11}$ showed fifty proton groups from the (d, p) reaction. This work, using 2-Mev deuterons, covered the region between the ground state and an excitation of 6.35 Mev in Al$^{28}$. The present work also covers this region and extends it upwards to neutron binding in Al$^{28}$. Each of the previously reported groups has been seen in the present work, and the very close agreement of the measured Q-values is a gratifying check on the conclusions of the earlier experiment and on the reliability of calibration of the new spectrograph. In addition to these fifty groups, approximately fifty additional ones have been measured. This work is virtually completed except for the calculation of small corrections caused by relativistic effects. The final Q-values will be listed in the next Progress Report.

As time permits, the plates exposed during the angular-distribution measurements on this reaction are being read to obtain information on states lying higher
VI. C1\textsuperscript{35}(d, p)C1\textsuperscript{36} AND C1\textsuperscript{35}(d, α)S\textsuperscript{33} REACTIONS

While C. H. Paris was associated with the group as a part of last summer's Foreign Student Summer Project, plates were exposed for these reactions, using the broad-range spectrograph. He has continued the counting and analysis of these plates at the University of Utrecht in The Netherlands. Several different bombarding energies were used to enable the correct assignment of the observed groups. While the analysis is still in progress, the results thus far indicate, for the C1\textsuperscript{35} (d, p)C1\textsuperscript{36} reaction, a ground-state Q-value of 6.43 Mev and excited states, in C1\textsuperscript{36}, at 1.16, 1.61, 1.95, 2.47, and 2.52 Mev. For the C1\textsuperscript{35}(d, α)S\textsuperscript{33} reaction, the ground-state Q-value is 8.27 Mev and excited states in S\textsuperscript{33} appear at 0.84, 1.97, 2.35, 2.87, and 2.94 Mev. A check on these values and assignments will be obtained when data are available from plates which have not as yet been counted. (C. H. Paris)

VII. ANGULAR DISTRIBUTIONS OF Ca(d, p) REACTIONS

Another series of runs on the Ca\textsuperscript{42}(d, p)Ca\textsuperscript{43} angular distribution was completed. The data now include measurements at 5-degree intervals from 10 to 60 degrees and at 10-degree intervals from 60 to 100 degrees; all at a deuteron energy of 7 Mev. The plates have been counted, and the angular distributions are being assembled. The attempt to fit the data with Butler curves has been continued.

Preliminary results indicate that the best fit to the maximum in the angular distribution corresponding to the Ca\textsuperscript{41} and Ca\textsuperscript{43} ground states for $l_n = 3$ is given when $R = 7.5 \times 10^{-13}$ cm, an even larger value for the radius than that mentioned in the previous Progress Report. The group corresponding to the 1.00-Mev state in Ca\textsuperscript{43} shows a distribution similar to that of the ground state. This was not expected. A state at 0.996 Mev, seen by Lindqvist and Mitchell\textsuperscript{12}, in the $\beta^-$ decay of K\textsuperscript{43} is reported as having parity opposite to that of the ground state.
The results on the first four states of Ca$^{41}$ are in general agreement with those of Holt and Marsham$^{13}$. However, the weak group produced by the 2.015 level in Ca$^{41}$ is resolved from the strong group from the 1.947 level. The former appears to show a distribution similar to that of the ground state, although the intensity is so low (10 percent of the ground state) that the shape is not well determined. (C. K. Bockelman and D. B. Guthe)

Note: The enriched magnesium and calcium isotopes mentioned in the foregoing report were obtained from the Stable Isotopes Division of the Atomic Energy Commission, Oak Ridge, Tennessee.

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PUBLICATIONS


THESSES


W. W. Buechner
I. THE FIRST EXCITED STATE OF POSITRONIUM

Further refinements in the timing circuits give a short time stability within 2-3 parts per 10^4 in the equality of light-on light-off intervals.

Excitation with the line in the tin spectrum 0.7 A. u. away from the Lyman absorption in positronium is being compared with excitation using the mercury spectrum under otherwise identical conditions. The strongest line in the mercury spectrum is \(\sim 100\) A. u. from the Lyman absorption and should yield no effect.

The counting rate change during excitation with the tin spectrum is \((0.11 \pm 0.08)\) percent with mercury \((0.0 \pm 0.09)\) percent. The experiment is still in progress. (H. W. Kendall and M. Deutsch)

II. ZERO-ZERO TRANSITION IN Ge\(^{72}\)

A delayed 680 kev conversion electron following the \(\beta\) decay of 14 hour Ga\(^{72}\) has been attributed\(^1\) to a spin zero to spin zero transition ending in the ground state of Ge\(^{72}\). This electron is observed in about 0.3 percent of the Ga\(^{72}\) decays.

We have measured the half life \(\tau\) of this state by delayed \(\beta\beta\) coincidence techniques and find \(\tau \approx 0.6\) \(\mu\)sec, somewhat longer than was reported earlier\(^1, 2\).

The conversion electron is also observed with delayed \(\beta\gamma\) coincidence, and experiments are under way to determine the energies of the \(\gamma\)-rays which precede it. (H. W. Kendall and M. Deutsch)

III. INTERNAL PAIR CREATION IN BETA DECAY

Using the apparatus, the construction and operation of which has already been described in previous LNS Progress Reports, it was found that the beta decay of a source of P\(^{32}\) obtained from Chalk River (\(\sim 25\) mcs in one milligram) was accompanied by electron positron pair creation with an abundance of \(0.9 \pm 0.6 \times 10^{-8}\).
Radioactivity Group

per beta decay. Due to an unresolved background effect (the fractional contribution to this number by external pair creation in the source material), no definite conclusions can at this time be reached regarding the absolute probability of internal pair creation. However, the experiment does indicate that the effect is probably not greater than $1.5 \times 10^{-8}$ per beta decay. (J. S. Greenberg)

IV. RAYLEIGH SCATTERING

A preliminary investigation has been made of the practicability of measurement of the Rayleigh scattering cross section of $\gamma$-rays. Various possible geometries have been considered and a few rough measurements completed. It has been found that the simpler methods of measurement require unshielded sources of $\gamma$-rays, the intensities of which are too high for a crowded laboratory.

Two other possible lines of investigation are being examined. The first is to measure the Rayleigh scattering of a collimated beam of $\gamma$-rays from a very strong source. The second is to confine our measurements to small angles, where the Rayleigh scattering cross section is relatively large, but it is very difficult to distinguish between Rayleigh and Compton scattered $\gamma$-rays.
(R. C. McCall)

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ADDRESSES


R. D. Evans
M. Deutsch
I. ALPHA PARTICLE EXPERIMENTS

Use of the 31 Mev alpha beam for the study of the inelastic $\alpha$ scattering process is continuing. Angular distributions have been obtained for the first and third excited states in the reaction $\text{Li}^6(\alpha, \alpha')\text{Li}^6^*$, (it is of interest that the second excited state $T_1$ is not observed), and for the first and second excited states in the reaction $\text{Cl}^{12}(\alpha, \alpha')\text{Cl}^{12^*}$.

The curves show well defined maxima and minima with no center of mass angular symmetry. The angular distribution of inelastic $\alpha$'s from the first excited state of $\text{Li}^6$, for example, has the first minimum and maximum at cm angles of 36° and 45°, respectively, with a peak to valley ratio of 3 to 1. The third excited state first minimum occurs at 43° and the maximum at 50° with a peak to valley ratio of 2 to 1. Both curves appear to peak strongly forward.

Measurements are being extended to forward angles to investigate the apparent forward maxima. A 20 channel pulse height analyzer is being constructed, (modified Oak Ridge model), to facilitate these measurements.

M. S. Livingston
I. OPERATION AND MAINTENANCE

The synchrotron pole pieces were removed and the doughnut removed to replace the rf cavity. As mentioned in the LNS Progress Report of November 30, 1954, the new cavity has a feed system going to the inner wall by drilling a hole for the rf probe through the cavity. We have been able to get a more efficient feeding system in this manner.

When the doughnut was replaced, the beam was not found at the previous settings of the field correction coils. In an endeavor to find the beam, we measured the impedance of each C magnet and the absolute magnetic field at injection (more specifically, the passage of the field through zero field). The magnetic field appeared reasonable enough to be corrected through the correction coils at injection; however, the betatron shunt impedance of the various C's were remeasured and corrected to be the same within a few percent.

The beam was finally obtained by setting the correction coils to give a maximum beam collection at the last doughnut section before the gun. By using an ejector coil at this section, it was then found that an appreciable fraction of the electrons hitting the target must have gone around at least one and a half times. These were detected by a scintillating crystal in the beam. By maximizing these electrons the betatron beam appeared.

The beam, when obtained, was not up to its former intensity. More measurements were made on the magnet impedances, and the betatron shim material was adjusted to equalize these. With the intensity still sub-normal, a beam contraction system was hooked up. It consisted of a hydrogen thyratron and resistor in series placed across the output of the betatron coil. When fired, it has the effect of turning off the betatron acceleration for a time given by L/R, where R is the resistance in the circuit and L is the inductance of the betatron coil. When put in operation, this circuit immediately increased the intensity to normal or a little above and stabilized the intensity. The feeling is that it is possible, by critical settings of the field correction coils, to establish an injection mechanism which prevents the beam from hitting the gun on subsequent turns after injection. How-
ever, if one has not found these settings, the contractor coil allows efficient and non-critical injection. This contraction scheme had been tried previously, similar to a manner suggested by Kerst, and its effect at that time was not significant.

The betatron to synchrotron capture is not as efficient as it had been previously. This effect seems to be connected with a non-linear betatron condition which in turn seems related to a non-linear hysteresis in the betatron shunt. This effect is being investigated. (W. Lobar, L. S. Osborne, G. E. Pugh, D. M. Ritson and A. Wattenberg)

II. COHERENT PHOTOPRODUCTION OF $\pi^0$ MESONS

New electronic circuits have been built using the same principles as the previous circuits. Instead of 6199 photomultipliers with a stage of amplification, 1P21 tubes are being tried directly. A multi-channel Garwin coincidence circuit has been built; four channels of this will be used (two Cerenkov counters, two plastic scintillation counters) which should give a faster coincidence resolving time than was possible previously.

A Bey coincidence circuit has been tried. The resolving time with pulses coming from two scintillation counters ($\gamma$ radiations of Co$^{60}$ on plastic or liquid scintillator viewed by a C7165A or 1P21 tube operated at 1600 volts) is at half maximum width of the order of $4 \times 10^{-9}$ sec. But the resolving time for delayed pulses coming from only one counter is lower, and of the order of $1.5 \times 10^{-9}$, and the delay between two pulses can be measured with an error inferior to $10^{-10}$ sec. Work is being done to improve the resolving time obtained with two counters and to reduce the fluctuation in delay between the two pulses by an appropriate shaping of the pulse. (G. Davidson, R. Barringer, L. S. Osborne and R. Meunier)

III. HIGH PULSE DETECTION BY SHORT PULSE

In the study of the momentum dependence of the $\pi^+$ meson excitation function by Dr. A. Winston, a deviation from the momentum law was found which might possibly be more than a statistical error. We intend to repeat the experiment in an attempt to check the reality of this deviation. We are now reassembling and testing the required electronics. (L. S. Osborne and B. Richter)
IV. PHOTODISINTEGRATION OF THE DEUTERON

We have continued on with the scanning of the exposures. An estimated 1800 tracks have been found. The plates have been double, and in some cases, triple-scanned as a check on the efficiency of scanning. The greater fraction of the tracks that leave one emulsion has been traced to the next emulsions in the stack. In the near future the results will be analyzed. (S-C. Fung, Nora Mohler, L. S. Osborne and D. M. Ritson)

V. BROOKHAVEN EMULSION EXPERIMENTS

The Brookhaven machine has been out of action and no new exposures have been made in the last three months. In the interval, plans have been made in conjunction with the Brookhaven group to obtain a much improved magnetically analyzed K⁺ beam. On the assumption that such a beam will be available when the machine starts up again, we have nearly completed preparations to handle 10" × 15" emulsion stacks in which K⁺ secondaries would be stopped with fair probability. We have built and tested stainless steel plate racks to develop twenty such plates at a time and believe we have the capacity to develop with ease 60 plates a week. We have converted one microscope satisfactorily to scan such plates and are in the process of converting a second. We are also constructing a microscope on which we can trace and perform precision measurements.

We have, in conjunction with Gerson Goldhaber, exposed plates at Berkeley.

As soon as we obtain suitable exposure conditions (suited to a small scanning group) we shall expose a large emulsion block. (S-C. Fung, Nora Mohler, L. S. Osborne, A. Pevsner and D. M. Ritson)

VI. DEUTERONS FROM HIGH ENERGY PHOTOEFFECTS

Several changes were made in the electronic part of the detecting arrangement which will improve the stability and accuracy. (H. Medicus)
VII. LIQUID HYDROGEN TARGET

The liquid hydrogen target has been completed and tested satisfactorily. We are currently engaged in installing the target and its control apparatus in position for use with the beam. (G. S. Janes, L. G. Hyman and C. J. Strumski)

VIII. GAMMA RAY SCATTERING BY A COMPLEX NUCLEAR SYSTEM

A study of the assumptions involved in relating the scattering of a nuclear system to the scattering of free nucleons is in progress. The problem is divided into two parts. First, the scattering of a free nucleon is related to the scattering of a bound nucleon. Second, the scattering of one bound nucleon is related to the scattering of many bound nucleons. The first is still in progress, but results on the latter problem have been obtained.

These results, although derived in a more general way, are essentially identical with those obtained by Waller and Hartree for electron scattering of X-rays. If one views the ground state of the nucleus \( \psi_0 \) as an anti-symmetrized product of single particle wave functions \( \varphi_i \), then the scattering of the complex system can be expressed in terms of the scattering by an individual particle in the state \( \varphi_i \).

If the individual scattering is given by

\[
\sigma_{\text{elastic}} = \left| \int \varphi_i a \varphi_i \, dv \right|^2
\]

\[
\sigma_{\text{inelastic}} = \sum_{f \neq 1} \left| \int \varphi_i a \varphi_f \, dv \right|^2
\]

If we number the possible single particle states 1 to N for those occupied, and N + 1 to \( \infty \) for the unoccupied states, then we obtain for the scattering of the complex system,

\[
\sigma_{\text{elastic}} = \left| \sum_{i=1}^{N} \int \varphi_i a \varphi_i \, dv \right|^2
\]
For high energies relative to the binding energy of the $\varphi_i$, the expression for $\sigma_{\text{inelastic}}$ reduces to

$$
\sigma_{\text{inelastic}} = \sum_{f=N+1}^{N} \left| \sum_{i=1}^{N} \left( \varphi_i a \varphi_f dv \right) \right|^2
$$

The Pauli exclusion terms make this less than $N$ times the single particle inelastic cross section. This reduction in elastic scattering may also be considered to result from a placement of nucleons in the nucleus which is more uniform than statistical because the anti-symmetric $\Psi$ prevents nucleons from coming really close together. Any strongly repulsive short range force existing further in the nucleus will accentuate this uniformity and further decrease the inelastic scattering. Alternatively, any strong short range attractive force will make the placement less uniform and will enhance the inelastic scattering.

The results so far observed for the inelastic scattering are fully compatible with the concept of random placement of the nucleons. (G. E. Pugh)

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B. T. Feld, "Nuclear Polarization Resulting from $\pi$-Meson Production", University of Illinois, Urbana, November 29, 1954.


PUBLICATIONS


L. S. Osborne
I. INVESTIGATION OF THE BOHR-MOTTELSON NUCLEAR MODEL

A. Individual Particle Levels in a Deformed Potential

The calculations of the approximate wave function for the motion of a single nucleon in a strongly deformed field are proceeding on a larger scale. With the help of the Whirlwind computer, the energy spectrum of a deformed field has been computed by taking into account the combinations of the states in different shells. Here states up to the occupation number of 150 have been considered. The energy is determined as a function of the deformation. Using the observed deformation as deduced from rotational spectra and Coulomb excitation, one can determine the properties of the lowest state of the nucleus by filling up the energy states with the right number of protons and neutrons. The states are different from the original spherical shell model. It turns out that the agreement with the observed spins and magnetic moments is much better with this deformed model. We, therefore, conclude that the fundamental idea of considering the nucleus as a system of free particles in a deformed potential well has approximate validity. (K. Gottfried and V. F. Weisskopf)

B. Derivation of the Hamiltonian by a Canonical Transformation

Collective modes of motion of the nucleus describing rotation, shape- and volume-vibration of the ellipsoid of inertia are introduced by means of a canonical transformation. In addition to the collective coordinates, "body-fixed" particle coordinates are introduced. Their range of variation is somewhat restricted; and this is taken into account by imposing suitable subsidiary conditions on the wave function. These subsidiary conditions greatly simplify also the expression for the kinetic energy operator, which becomes the sum of rotational, vibrational and intrinsic kinetic energies.

The Hamiltonian thus obtained resembles closely the one derived by Bohr on the basis of the hydrodynamic model\(^1\). The moments of inertia in the rotational energy term are identical with Bohr's. One difference is the appearance of terms depending explicitly on the number \(A\) of particles in the vibrational energy. (F. Villars)
C. Rotational Spectrum in the Bohr-Mottelson Theory

It is well known that the Bohr-Mottelson theory gives a level spacing of the rotational terms that is much too wide, if the moments of inertia are derived from the intrinsic quadrupole moments. An investigation is in progress to analyze this inconsistency. It appears now that it is due to the neglect of certain perturbation terms in the rotational energy, which, by second-order perturbation theory, produce a very large correction to the rotational spectrum. (F. Villars)

II. DETERMINATION OF THE IMAGINARY POTENTIAL WITHIN THE NUCLEUS

An attempt was made to estimate the imaginary potential which is introduced into the optical model of the nucleus in order to describe the compound nucleus formation. This estimate is made on the idea that compound nucleus formation is due to elastic collisions of the incoming particle with one of the constituents inside the nucleus. These constituents are considered to be part of a Fermi gas of free particles within a potential well. Calculations were made on this basis and give an imaginary potential whose energy dependence is reproduced in Fig. 11.1. The agreement with the present experimental results is surprisingly good. (A. M. Lane and C. F. Wandel)

III. THE COMPOUND NUCLEUS

A general critical review of the concept of the compound nucleus and its formation has been compiled. (F. L. Friedman and V. F. Weisskopf)

IV. NUCLEAR RADIUS AND NUCLEAR FORCES

This work, reported in preceding LNS Progress Reports, has been completed and the following abstract has been submitted for the Washington Meeting of the American Physical Society.

The difference between the "radius" of the nuclear matter distribution and the nuclear force radius \( r_n \sim 1.45A^{1/3} \times 10^{-13} \text{ cm.} \) for \( A \geq 50 \) is interpreted
Figure 11.1

Imaginary part of complex potential as function of neutron bombarding energy.
Theoretical Group

as a consequence of the finite range of nuclear forces. Assuming that the matter distribution coincides with the charge distribution as determined at Stanford, \( r_e \sim 1.2\lambda^{1/3} \times 10^{-13} \text{ cm} \), we sum up the nuclear interactions of an incident nucleon for various proposed internucleon potentials, \( V(r) \). We also evaluate induced spin, matter, and charge polarizations of the nuclear matter due to an impinging nucleon as a test of the convergence of these calculations. The aim here is to see what ranges and strengths of \( V(r) \) are appropriate, say, to neutralize the Coulomb barrier for protons at a distance \( \sim r_n \). This approach is sensitive, in first order, only to the long-range behavior of the spin and charge independent portion of \( V(r) \). It leads to the following conclusions: Either the neutron distribution extends at least 10 percent beyond the proton one or \( V(r) \) must contain a sizeable amount of Wigner interaction with the range of a meson Compton wavelength. Such an interaction is not found in recent meson theoretically deduced \( V(r) \). This result is insensitive to details of the tail of the charge distribution. 

(S. D. Drell)

V. PROTON BREMSSTRAHLUNG

The differential cross section for proton bremsstrahlung is calculated for low energy X-rays emitted by \( E = 2 \text{ Mev} \) protons which scatter in the field of tin nuclei \((Z=50)\). This is essentially a classical problem of the dipole radiation from a proton moving in a Coulomb field since the proton's deBroglie wavelength is much shorter than the distance of closest approach to the nucleus. However, the classical orbit calculation for X-rays emitted at 90° to the proton beam in a 3 kev energy interval about \( \hbar \omega = 150 \text{ kev} \) predicts a differential cross section, \( \sigma_c(90^\circ) = 2.1 \times 10^{-31} \text{ cm}^2 \), in disagreement with the experimental result of H. Mark, \( \sigma_{\text{exp}}(90^\circ) = 1.3 \pm 0.5 \times 10^{-31} \text{ cm}^2 \). Since corrections due to shielding by the orbital electrons and due to higher multipoles are negligible, this discrepancy must be due to quantum corrections resulting from the energy loss of the proton to the radiation. We have expanded the exact quantum-mechanical matrix element for dipole bremsstrahlung, as derived by Sommerfeld, in a power series in \( \hbar \omega/E \). Keeping the zero and first-order terms, we have carried out the necessary integrations. The correction thus obtained serves to lower the calculated cross section by 25 percent and to establish agreement with experiment. This work has been completed and prepared for publication. (S. D. Drell and K. Huang)
VI. INTERNAL PAIR-PRODUCTION ACCOMPANYING $\beta$-TRANSITIONS OF NUCLEI

The ratio $N^+/N^-$ of the number of positrons to the number of electrons as well as the energy spectrum of the positrons in the internal pair-production accompanying $\beta$-transitions of nuclei were first investigated theoretically by Arley and Møller in 1938. The preliminary experimental results of J. Greenberg, however, seem to disagree with the quoted theoretical treatment. Since the model used by Arley and Møller—that of a single particle transition in the nucleus—did not appear to be justified on a priori theoretical grounds, and certain questionable approximations were introduced in arriving at their final answer, it seems worth while to re-examine this problem.

Apart from the pair-production by the $\beta$-particle from the decay (Møller scattering of an electron in the negative energy sea), the nucleus in its ground state is pictured to create first the pair purely by its Coulomb field, with the whole nucleus recoiling to conserve momentum, while the internal nuclear state remains unchanged. The nucleus then undergoes $\beta$-decay, thus conserving energy in the whole transition. (The pair-production is thus effected by the same mechanism as vacuum polarization.) Summing up the matrix element for this process and one in which the $\beta$-decay occurs before pair production, one obtains a matrix element that is $Z$-independent ($Z=$ nuclear charge). Hence the pair-production is essentially due to the single proton that is transformed from a neutron in the $\beta$-decay, thus justifying the model used by Arley and Møller. For an allowed $\beta$-decay, the ratio $N^+/N^-$ and the positron spectrum are independent of the type of $\beta$-coupling used. Therefore, the formulas given by Arley and Møller, who used a scalar coupling for the $\beta$-decay, are correct, and discrepancy with experiment may be due to the approximation used in their numerical work. A more exact calculation is under way. Internal pair-production accompanying first forbidden $\beta$-transitions is also being considered. This is perhaps theoretically a more interesting case, as the results for this case will depend on the type of $\beta$-coupling used. (K. Huang)

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Theoretical Group

4. J. Greenberg, private communication.

PUBLICATIONS


V. F. Weisskopf
APPENDIX A

PERSONNEL LISTING BY PROJECTS

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