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RADIATION LABORATORY

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SUMMARY OF RESEARCH PROGRESS MEETING OF OCTOBER 25, 1951

Sergey Shewchuck

December 11, 1951

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Berkeley, California

SUMMARY OF RESEARCH PROGRESS MEETING OF OCTOBER 25, 1951

By S. Shewchuck

Radiation Laboratory, Department of Physics
University of California, Berkeley, California

December 11, 1951

I. Heat of Reaction and Crystal Energy in High Temperature Hydrolysis of Some Solid Trichlorides. By B. B. Cunningham.

A general type of reaction with H_2O vapor at high temperature is:

$$MCl_{3(solid)} + H_2O_{(gas)} \longrightarrow MOCl_{(solid)} + 2\ HCl_{(gas)}$$

where M is any of the rare earth, transuranium or other elements which undergo this reaction. This reaction is used, as an example, in determining thermodynamic constants for unknown substances. The determination of thermodynamic properties is not easy, although there is now available accurate data of thermodynamic constants for many compounds. It is the business of chemists to know what reactions can take place and under what circumstances. Hence, knowledge of thermodynamic constants is indispensable. In the above reaction the thermodynamic properties of all compounds were known except for MOCl. Oxychlorides like this have a considerable nuisance value. The chemist then would like to be able to predict when such compounds will be produced in a given reaction.

An explanation of the method of determining the thermodynamic constants was discussed in theory employing the three laws of thermodynamics and expressions for the enthalpy and entropy are derived from the heats of reaction and the temperature. For a reaction to proceed spontaneously the entropy must always increase. Thermodynamics provides no information as to the reaction rates and

thus one cannot predict spontaneity of reaction. A new criterion of free energy change ΔF is required here, which is related to the equilibrium constant K.

For spontaneous reaction $\Delta F = \Delta H - T\Delta S < 0$

In the case of the oxychlorides

$$K = \frac{(MOCl)}{(MCl_3)} \times \frac{p_2\,(HCl)}{p_1\,(H_2O)}$$

where the relative effective concentrations of the components are used. But since MOCl and MCl_3 are solids and their concentrations are equal to one, then K is the ratio of HCl at pressure p_2 to H_2O at pressure P_1. Hence, the heats of reaction obtained from K at various pressures will give the thermodynamic properties of the oxychlorides.

In principal, one may get this information in other ways, as from ionic and crystal structure, by distances between charges and work energy needed to bring them into position in the lattice. From the position of the assemblage of charges one should be able to calculate the energy of the hypothetical separation of charges; $MCl_3 \rightarrow M^{+3} + 3\,Cl^-$. Also, it is known how much energy is required to get M^{+3} from the metal and 3 Cl^- from chlorine in the standard state. These calculations are reasonably good for simple ionic substances, e.g. 2 percent agreement for NaCl; but it cannot be carried out accurately for complicated crystals due to compound bonding. The crystal energy U_o is calculated from the formula as follows:

$$U_o = 279\,\alpha^2 \left(\frac{\ell}{M}\right)^{\frac{1}{3}} A\,\delta_o \left(1 - \frac{1}{n}\right) \quad \text{K cal/mole}$$

largest common factor in valences of ions — radius of sphere — Madelung constant — repulsion term between crystals

$n = \frac{1}{2}$ is a close value for most crystals.

The values of crystal energy are found as follows:

for $LaCl_3$, U_o = 1124 K cal

for LaOCl, U_o = 1294 K cal

The lattice dimensions are known for a number of other oxychlorides. One can calculate the change in heat of reaction as one substitutes another metal for La in the reactions. In the change of La to Gd there is found to be an increase in U_o of 17.4 K cal for trichlorides and of 24.5 K cal for oxychlorides. The agreement is good in some cases when compared with the experimental measurement. The trichloride crystals are hexogonal and the oxychloride tetragonal.

II. Nuclear Momentum Distribution. By J. Cladis.

When protons are scattered with light nuclei such as C, D_2 and H_2, single collisions with the nucleons of these nuclei are possible, and this is manifested in the energy spectrum of the scattered protons. A 35 channel magnetic particle spectrometer is used to study the momenta distribution of the scattered protons at the time of collision and to try to set limits on internal momenta. The schematics of the apparatus is shown in Fig. 1. A four fold coincidence is used on the counters. The energy resolution of the spectrometer has been improved over the first version. Energy widths of the various components were computed. The resolving power was obtained by folding the energy widths together, and a check was obtained from the scattered protons of the H_2 target. The principal limitation to the resolution was the width of the GM counters. The GM tube array was moved four feet farther back from the magnet, and this improved the resolution by a factor of four. Also, the slit counter was decreased from ½ inch. to 1/8 inch., and a thinner window of 1 mil aluminum was used in the proportional counters. Overall resolution is consistent

so that one gets maximum counting rate for the resolution used. Fig. 2 gives the theoretical resolving power curve of spectrometer averaged over beam energy spread.

Fig. 3 gives the data on the energy spectrum obtained at 30° scattering angle of protons from C, H_2 and D_2. Pulse height analysis has been done to show that the scattered particles observed are really protons. The targets used were water, heavy water, polyethylene and carbon. The data from O_2, not included in the graph, also gives a curve consistent with its being 16/12 of the data from C.

Fig. 4 gives the data at a scattering angle of 40°. The spectra are more accurately described since the energy channels are narrower at this angle. Also, a comparison is made with the theoretical curves obtained by using Gaussian and Chew-Goldberger momentum density distributions. The peak energies of the protons from deuterium and carbon are less than the corresponding peaks from hydrogen.

Fig. 5 gives the data on the ratio of the number of protons from deuterium as compared with hydrogen. The total number of the scattered protons is used for this graph, plotted against the angle. The curve drawn through these data has the same form as that of the n,p differential cross section as a function of the angle. At 80° in the center of mass system the observed ratio is 1.1; it should be $\frac{\sigma_{n,p} + \sigma_{p,p}}{\sigma_{p,p}} = \frac{0.7 + 3.8}{3.8}$, or 1.18^+, if the scattering were due entirely to the free nucleon - nucleon collisions.

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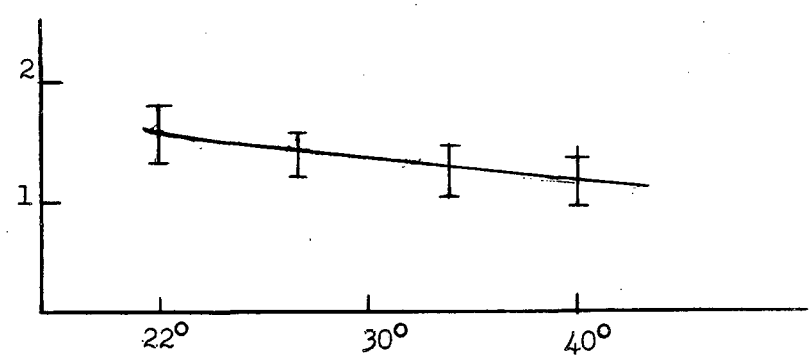

Fig. 5

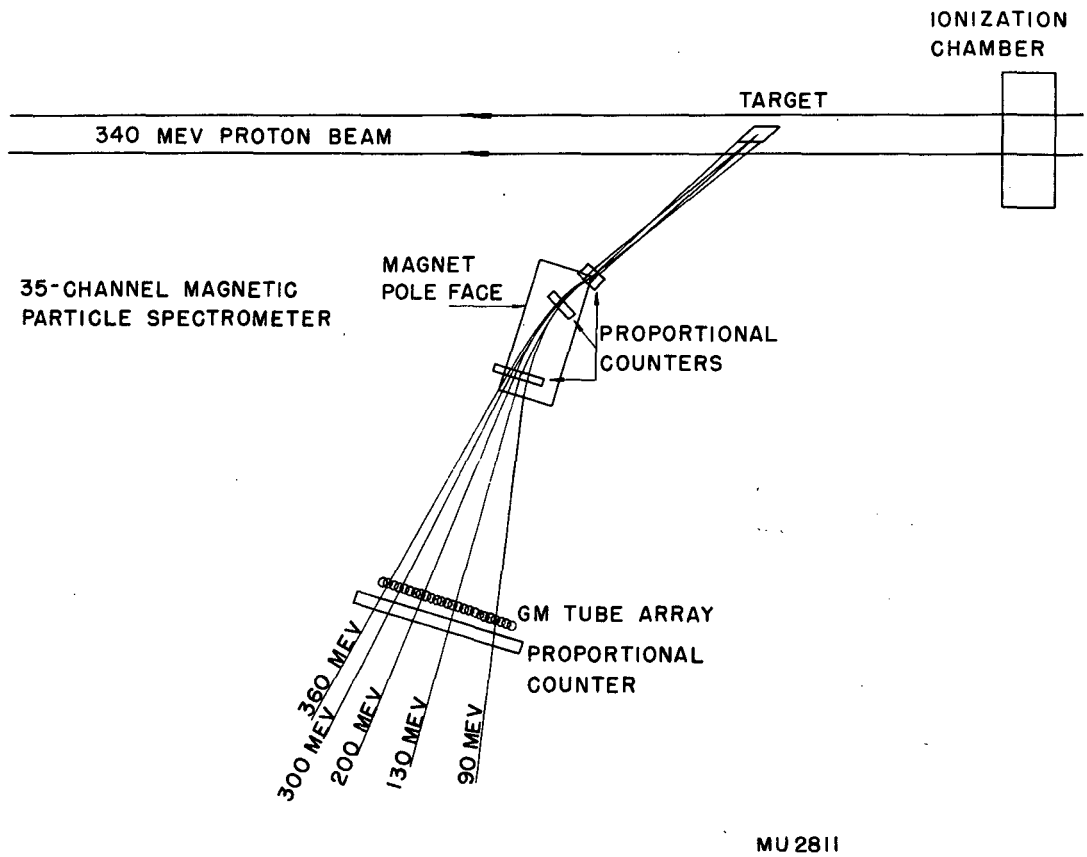
IONIZATION
CHAMBER
TARGET
340 MEV PROTON BEAM
35-CHANNEL MAGNETIC
PARTICLE SPECTROMETER
MAGNET
POLE FACE
PROPORTIONAL
COUNTERS
GM TUBE ARRAY
PROPORTIONAL
COUNTER
360 MEV
300 MEV
200 MEV
130 MEV
90 MEV
MU 2811

Fig. 1

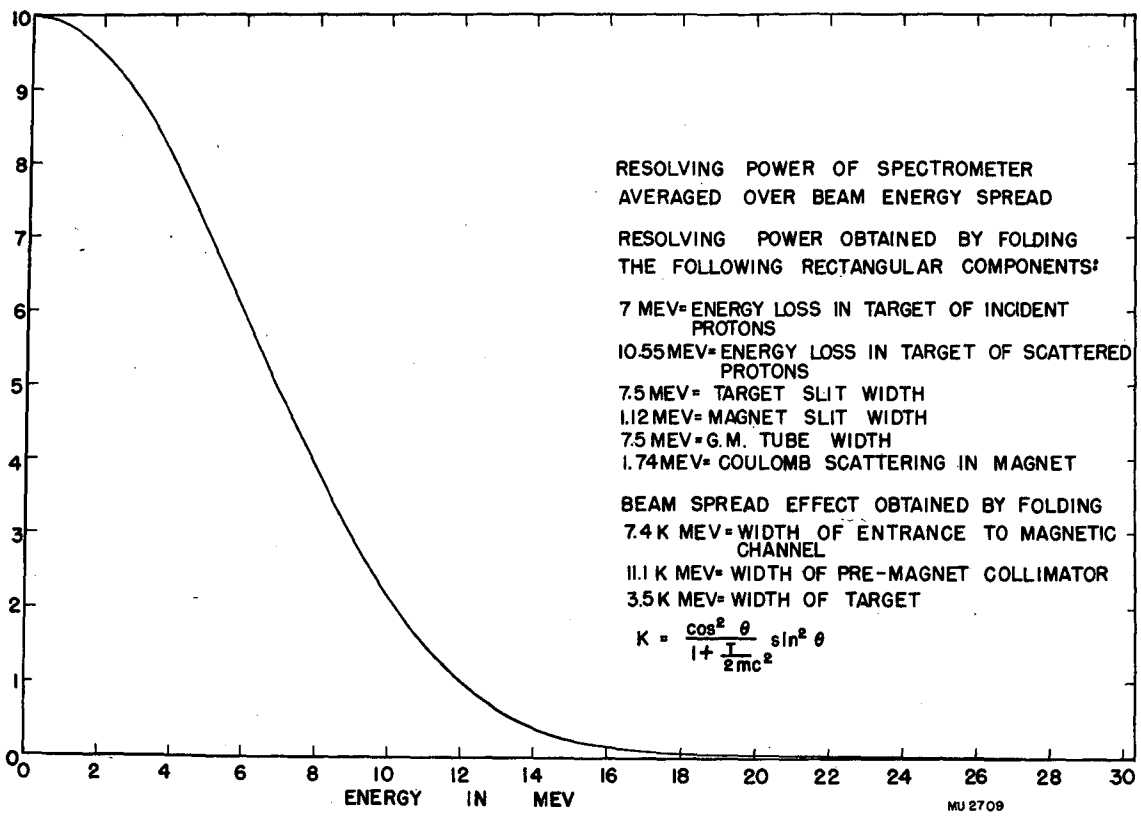
RESOLVING POWER OF SPECTROMETER
AVERAGED OVER BEAM ENERGY SPREAD
RESOLVING POWER OBTAINED BY FOLDING
THE FOLLOWING RECTANGULAR COMPONENTS:
7 MEV= ENERGY LOSS IN TARGET OF INCIDENT PROTONS
10.55 MEV= ENERGY LOSS IN TARGET OF SCATTERED PROTONS
7.5 MEV= TARGET SLIT WIDTH
1.12 MEV= MAGNET SLIT WIDTH
7.5 MEV= G.M. TUBE WIDTH
1.74 MEV= COULOMB SCATTERING IN MAGNET
BEAM SPREAD EFFECT OBTAINED BY FOLDING
7.4 K MEV= WIDTH OF ENTRANCE TO MAGNETIC CHANNEL
11.1 K MEV= WIDTH OF PRE-MAGNET COLLIMATOR
3.5 K MEV= WIDTH OF TARGET
$K = \frac{\cos^2\theta}{1+\frac{T}{2mc^2}} \sin^2\theta$
ENERGY IN MEV
MU 2709

Fig. 2

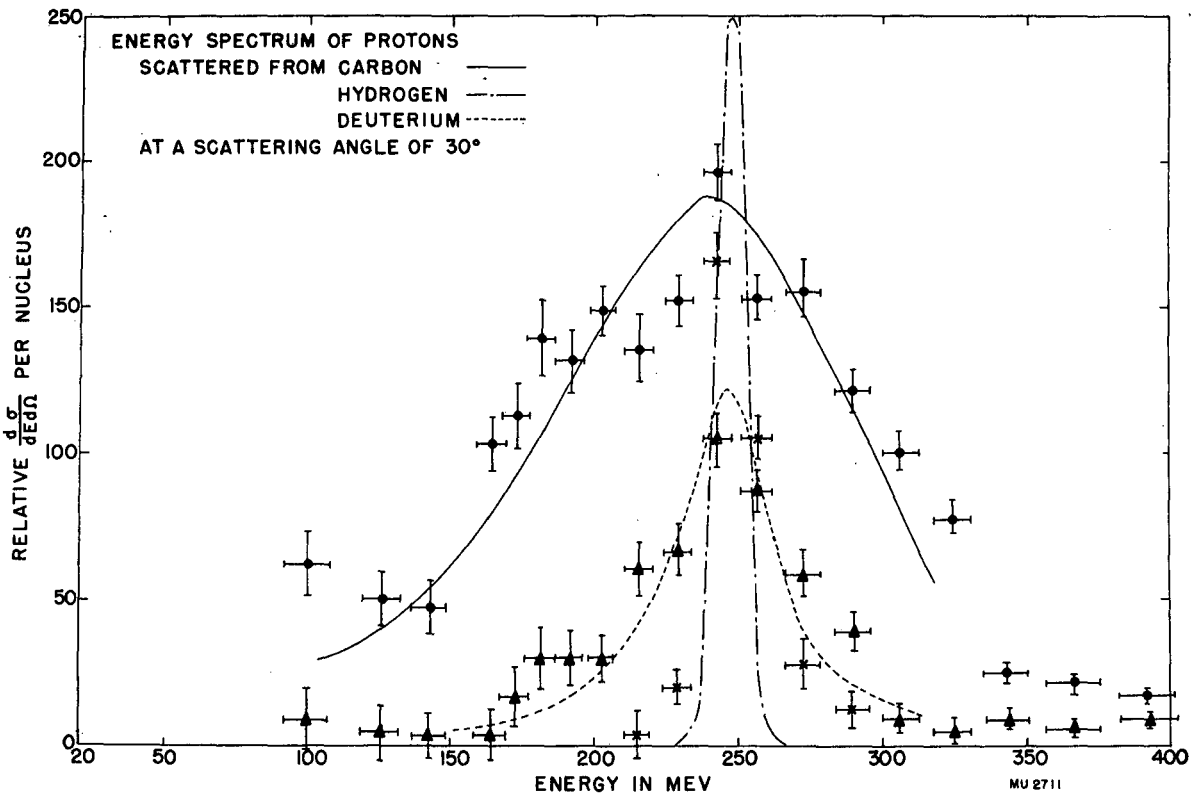
ENERGY SPECTRUM OF PROTONS
SCATTERED FROM CARBON
HYDROGEN
DEUTERIUM
AT A SCATTERING ANGLE OF 30°
RELATIVE $\frac{d\sigma}{dE d\Omega}$ PER NUCLEUS
ENERGY IN MEV
0
50
100
150
200
250
20
50
100
150
200
250
300
350
400
MU 2711

Fig. 3

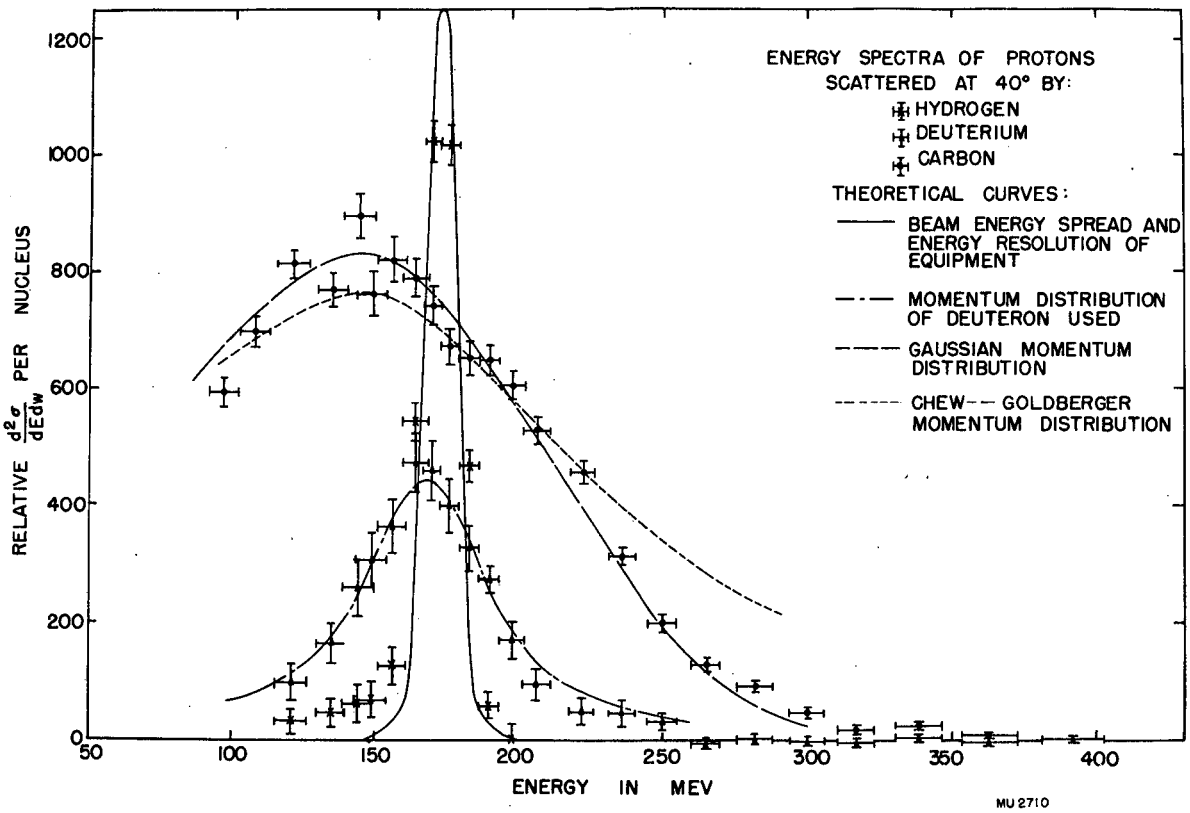
ENERGY SPECTRA OF PROTONS SCATTERED AT 40° BY:
HYDROGEN
DEUTERIUM
CARBON
THEORETICAL CURVES:
BEAM ENERGY SPREAD AND ENERGY RESOLUTION OF EQUIPMENT
MOMENTUM DISTRIBUTION OF DEUTERON USED
GAUSSIAN MOMENTUM DISTRIBUTION
CHEW-- GOLDBERGER MOMENTUM DISTRIBUTION
RELATIVE $\frac{d^2\sigma}{dEd\omega}$ PER NUCLEUS
ENERGY IN MEV
0
200
400
600
800
1000
1200
50
100
150
200
250
300
350
400
MU 2710

Fig. 4