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Charged Particles from the 14-Mev Neutron Interaction with Zirconium*†

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This experiment was performed in order to investigate the mechanism of the zirconium (n,p) and (n,np) reaction at 14 Mev. The energy distribution of particles interpreted as protons is found to peak at about 2 Mev. The cross section for charged particle emission was found to be 87 ± 22 mb. The angular distribution below 8 Mev is consistent with the compound nucleus model, but above 8 Mev a forward peak is observed.

I. INTRODUCTION

WHILE the theory of direct interactions proceeding to definite final states has been fairly well developed in recent years,[1-3] only a small amount of work[4] has been done with reactions proceeding to ill-defined or overlapping states. The statistical compound nucleus model[5] has been successfully applied to the low-energy products of fast particles with the heavier nuclei, but there is considerable evidence for direct interaction in the production of the high-energy products. This is seen from the angular distribution of the high-energy products. Direct interaction theory predicts a forward peak which has been found by inelastic neutron scattering experiments,[6,7] and in several (n,p) reaction studies.[8-10]

In addition, compound nucleus theory predicts a Maxwellian energy distribution which has generally been observed, although usually with more high-energy particles than expected. But the peak of the distribution, in the case of the (n,p) reaction, has been seen to be at a lower energy than predicted by the original theory, possibly indicating higher Coulomb barrier penetration. The total cross section is also generally higher than predicted.

The present experiment is an attempt to add to the body of data from which it is hoped a suitable theory may be constructed. It can also serve to test a theory due to Kikuchi[11] which introduces a diffuse nuclear potential into the energy distribution calculation. The results of the experiment can also be compared with previous work in inelastic neutron scattering from zirconium.[7] It is the first of a proposed series of such experiments, the second of which, with Co^{59}, is now being analyzed.

II. EXPERIMENTAL TECHNIQUE

A. Apparatus

A tritium gas target with cooled foils[12,13] was used with the Northwestern University electrostatic accelerator as a neutron source. Neutrons emerging at 90° to the deuteron beam were used.

A nuclear emulsion plate camera, similar to one built by Rosen[9] at Los Alamos, was used. The lid of the camera was lined with 0.015-in. gold, and 0.007-in. platinum windows were provided for the neutron beam. The neutrons were collimated by a collimator built by Wood and Singletary[14] from Rosen's plans. The camera was shielded from scattered neutrons by paraffin. The camera was built with plates every 15°. The emulsion plane of each plate was raised 14° above the horizontal, to make the incidence of the protons closer to grazing. This made it possible to use thinner emulsions than with flat plates, without requiring that the target be close to the base plate of the camera. The plates were $\frac{1}{2}$ in.$\times$1 in., and 6 in. away from the base of the target support, as close as was feasible, in order to obtain the maximum solid angle per unit plate area. The camera was mounted directly to the collimator, which, with the paraffin shield and the camera vacuum system, was mounted on a cart, facilitating alignment to the neutron source.

The zirconium target, made from commercially available foil, 11 mg/cm^2, was mounted on a 0.007-in. platinum disk, supported by platinum wire.

400 μ Ilford *G*-special emulsion was used, in order to minimize the silver layer on the surface, and for the same reason was cut to size at this laboratory, since the pressure of the packing on the ends of the plate increases this surface layer greatly. The plates were developed by the conventional two-solution

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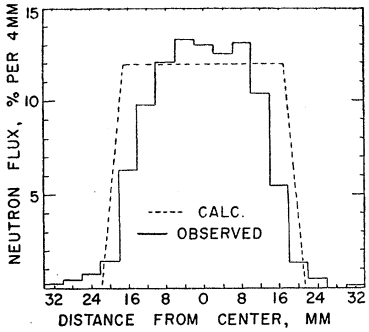

FIG. 1. The neutron beam profile, as seen at the sample target.

method,[15] and then soaked in wood rosin to decrease shrinkage.

To determine the neutron flux, neutron spectrum, and beam profile, an exposure was made in which a 400 micron plate was placed in the camera in the target position. The track count as a function of position was made to determine the beam profile, shown in Fig. 1. The neutron spectrum and flux for the exposure were determined by measuring the tracks in the forward direction falling into a 20° square prism in a measured volume of emulsion. The hydrogen density of the emulsion prepared at 50% relative humidity was taken as 0.052 g/cm^3. The resulting neutron spectrum is shown in Fig. 2.

To measure the relative flux for the various exposures, zirconium disks were placed in a reproducible position about 7 cm from the tritium gas target. Use was made of the $\mathrm{Zr}^{90}(n,2n)\mathrm{Zr}^{89}$ reaction,[16] which has a threshold of approximately 12.5 Mev. Coincidence counts of the annihilation gammas following the positron decay of the Zr^{89} thus determined the relative flux of fast neutrons.[17] As a confirmation of the relative flux values and for continuous monitoring, two long counters were also used in the various exposures. The agreement between the relative time integrated flux values as determined by the two methods was satisfactory. The long counters indicated that the flux was reasonably constant during the exposures.

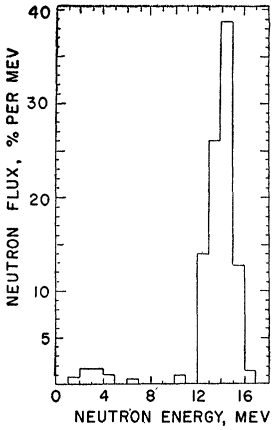

FIG. 2. The neutron spectrum, as seen at the sample target.

A background exposure was first made, with a time-integrated neutron flux of 5.71×10^8 neutrons/cm^2, followed by an exposure with a zirconium target with a flux of 7.76×10^8 neutrons/cm^2.

B. Data Analysis

The plates were scanned with binocular microscopes at about 1000 power. Only tracks beginning no more than 3 microns from the surface were accepted. In addition, their angles were required to be such that they could have come from the target. The dip angle was determined by measuring the depth of the track at the end of the first 30 microns, and the projected angle in the emulsion plane was measured with an eyepiece goniometer. The projected length of the track was measured either with an eyepiece reticle or a dial gage, depending on the length of the track. The dip of a number of long tracks was measured at several points to make sure that there was no significant distortion in the emulsion. 1000 acceptable tracks were found in the zirconium exposure, and 350 in the background exposure.

An attempt was made to determine what fraction of the tracks were produced by alpha particles for the range from 30 to 110 microns by grain and gap counts. No clean separation was possible and the results were ambiguous.

The dip was corrected for shrinkage, and the true range of the track was calculated, as was the angle with respect to the neutron beam and that with respect to the normal to the target. The proton energy was obtained from the range, using Barkas'[18] range-energy relation. The distance traversed by the proton in the target was calculated, assuming that it started in the central plane of the target. From this the energy of the proton was corrected for target thickness by the energy-

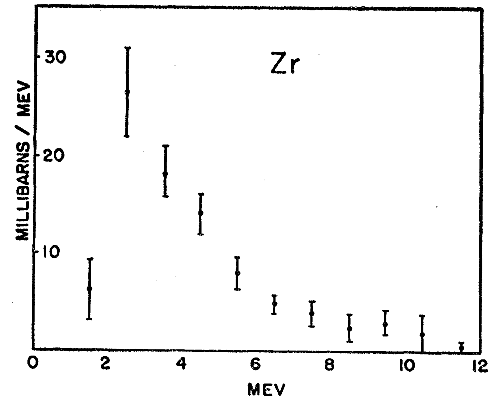

FIG. 3. The experimental energy distribution.

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loss equation of Livington and Bethe,[19] and from the mean excitation potential I from an experiment by Bichsel *et al.*[20]

The solid angle of each plate with respect to the target was calculated from the geometry, and the results of this calculation were compared to the results of an experimental determination, made by placing a U^{238} alpha source in the place of the target and counting the alphas in the plates.

The tracks were then divided into appropriate energy and angular intervals, and differential cross sections calculated. The background, multiplied by 1.36 (the ratio of the neutron fluxes in the two exposures) was then subtracted from the results of this signal-plus-background exposure.

Finally, the differential cross sections were transformed into center-of-mass coordinates by standard methods.[21]

III. RESULTS AND DISCUSSION

It was attempted to fit the data (shown in Fig. 3) with the statistical model prediction[5]:

$$N(E)=\mathrm{const}P(E)E\exp\{2[a(E_{\mathrm{ex}}-E_{\mathrm{kin}}-B)]^{\frac{1}{2}}\},$$

using barrier penetration factors $P(E)$ calculated both from a square well and a diffuse potential suggested by Kikuchi[11]:

$$V(r)=\frac{Ze^2}{r}-V_0\left[1+\exp\left(\frac{r-R}{c}\right)\right]^{-1}.$$

However, even assuming an admixture of alpha particles, it was impossible to make a reasonable fit. The low-energy peak in this energy distribution could also result from the (n,np) reaction, which is energetically possible in that region. However, attempts to fit the data with calculations of proton energy distributions from the (n,np) reaction using the statistical model have not been satisfactory.

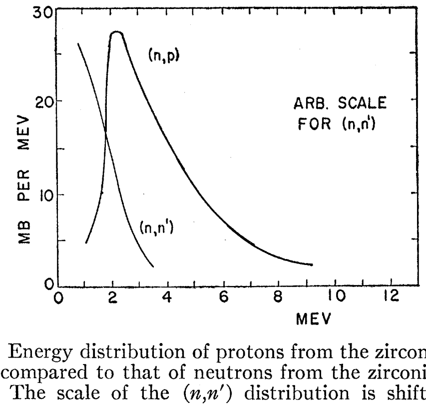

FIG. 4. Energy distribution of protons from the zirconium (n,p) reaction compared to that of neutrons from the zirconium (n,n') reaction. The scale of the (n,n') distribution is shifted by an arbitrary amount for comparison.

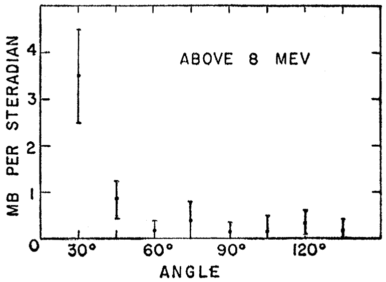

FIG. 5. The angular distribution of protons emitted with energies above 8 Mev.

Kikuchi of the University of Minnesota has estimated[22] from statistical theory and a diffuse potential that the upper limit of $\sigma(n,np)$ for Zr^{90} is about 30% of $\sigma(n,n')$ and that the cross section should be negligible from the other isotopes. This makes it about 15% of $\sigma(n,n')$ from all isotopes. He also estimated that the upper limit of $\sigma(n,p)$ is 9% of (n,n') for all isotopes. Using 410 mb for (n,n') from reference 7, $\sigma(n,np)\leqslant 62$ mb, and $\sigma(n,p)\leqslant 37$ mb. The sum of these, 97 mb, is to be compared with 87 mb observed in this experiment if $\sigma(n,\alpha)$ and $\sigma(n,d)$ can be neglected, as predicted by statistical theory.

The energy distribution peaks at an energy higher than that for neutron inelastic scattering[7] (Fig. 4) as was expected. The work of Colli *et al.*[23] does not show this peak, since their method only obtained a spectrum down to 3 Mev.

Figure 5 shows the angular distribution above 8 Mev, where a forward peak can be observed. This peak is evidence for direct interaction. About 10% of the particles produced in the reaction when interpreted as protons are above 8 Mev.

Wolfenstein[24] has shown that symmetry about 90° is expected in the angular distribution resulting from a compound nucleus reaction, which implies that the

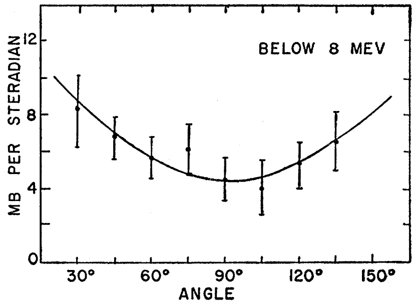

FIG. 6. The angular distribution of protons emitted with energies below 8 Mev. The curve is a least squares fit to a quadratic.

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results in Fig. 6, the angular distribution below 8 Mev, are consistent with this model.

The total cross section for charged particle emission was found to be 87±22 mb, which may be compared with the results of Armstrong and Brolley.[25] Calculated values obtained by the method of Blatt and Weisskopf,[5] give 8.9 mb for a square well. This result suggests that the diffuse nuclear potential is probably a better approximation to the truth than the square well.

IV. ACKNOWLEDGMENTS

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Multiple Scattering Correction for Proton Ranges and the Evaluation of the *L*-Shell Correction and *I* Value for Aluminum

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Multiple scattering corrections to proton range-energy measurements are discussed. Curves are plotted which give the fractional transmission of protons through a finite thickness of stopping material as a function of the initial proton energy and for various values of the ratio of the straggling and multiple scattering parameters. An application of these results to particular experimental situations shows that the Molière multiple scattering distribution gives a nearly correct representation of the experimental data on transmission and that a simple exponential distribution is not satisfactory. The case of straggling in gold is discussed in detail. Application of the results to the experimental range-energy relation in aluminum for protons of energy lying between 1 and 20 Mev is made. The experimental ranges can be described consistently with a mean excitation potential $I=163$ ev and with reasonable values of the L-shell binding energy correction.

I. INTRODUCTION

IN order to compare the results of range-energy measurements with the theory of stopping power, we must be able to make an accurate determination of a number of small quantities which appear as corrections in both the theory and the measurements. Failure to make a consistent application of such corrections in the past is partially responsible for the fact that stopping power relations based on a velocity-independent mean excitation potential have failed to predict correctly the measured ranges as a function of the energy of the penetrating particle. The discrepancies are small, but they are systematic and significant.

The most important correction to be made is in the stopping power relation itself. This is the correction for the binding energy of atomic electrons which has been discussed at some length by Walske.[1] Since, however, the present state of the theory of binding energy corrections is such that adjustable parameters should be introduced, a consistent application of binding energy corrections is hardly possible unless other required experimental corrections are made at the same time.

The most important of these other corrections is the multiple scattering correction. With the increasing accuracy of range-energy determinations it is of some importance that this correction be taken into account correctly. This will be the case even when the multiple scattering correction is of the same order as the error of measurement. The monotonic nature of the multiple scattering correction tends to remove a systematic trend in the discrepancy between theory and experiment. A more consistent application of the theory will then be possible. An immediate result of such a comparison of theory and experiment is a better determination of the mean excitation potential I.

We will be concerned here both with the method of determining the needed stopping power and range-energy corrections and with the application of these ideas in particular cases. The theory of the multiple scattering correction will be discussed in Sec. II. This section will contain also the results of numerical calculations; in particular, the transmission curves for protons of intermediate energy in several media. In Sec. III we make an application of these results to the

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* Now at the University of Southern California, Los Angeles, California, where part of this work was done.

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