Interaction of 14-Mev Neutrons with Deuterons*

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By coincidence counting of recoil deuterons in a proportional counter telescope the angular distribution for $n-d$ scattering has been measured using monoergic neutrons of 14-Mev energy. The measurements covered the angular range corresponding to neutrons being scattered at angles between 180° and 70° in the center of mass system, and by extrapolation an approximate curve was obtained over all angles. Upper limits can be placed on the probability of the n-$2n$ disintegration process for ejection of protons in the angular range from 0° to 120° in the center of mass system.

INTRODUCTION

THERE have been numerous investigations, both experimental and theoretical, on the interaction of fast neutrons with deuterons. The difficulty in the development of adequate theory for this three particle system has more or less paralleled the difficulty of experimenters in the determination of reliable data. Recent[1] and forthcoming data in the region of 100 Mev has encouraged more theoretical investigation.[2,3] Extensive calculations in the energy region below 14 Mev have been made by Massey and Buckingham,[4,5] who have compared the results of their calculations with experimental data on angular distributions for scattering[6] at 2.5 Mev and with data on total $n-d$ cross sections[7] at several energies up to 13.5 Mev. Recent cloud chamber data of Darby and Swan[8] at 2.5 Mev agrees in character, but disagrees quantitatively, with ion chamber data of Coon and Barschall.[6]

Angular distribution data have not, to our knowledge, been published for neutron energies greater than 2.5 Mev.[9] For neutron energies above the threshold ($\frac{3}{2}\times 2.2$ Mev) for the disintegration of the deuteron, any experimental search should seek to isolate the scattering process from the $n-2n$ process. Ageno *et al.*[7] give uncertain experimental evidence for the existence of the $n-2n$ process at 14 Mev and compare their results with those of other experimenters and with theory, though the information is extremely meager.

The experiments of Ageno *et al.* were made difficult by their not having an intense source of monoergic neutrons. The recent development[10,11] of sources in this laboratory using the reaction $T(d,n)He^4$ makes available an intense monoergic neutron source in the energy region from 12 to 18 Mev and simplifies the techniques required for measuring angular distributions and for differentiating between the scattering and $n-2n$ processes. In the present measurements, by using this source at a single neutron energy of 14 Mev, we have measured the angular distribution for scattering, and have been able to set upper limits for the $n-2n$ reaction over the angular region, for proton emission, from 0° to 80° in the laboratory system.

APPARATUS

The triple coincidence counting telescope and associated equipment used by Barschall and Taschek[12] for measuring $n-p$ scattering has been used, with methods similar to theirs, to investigate $n-d$ scattering. For most of the details regarding the neutron source, the proportional counter telescope, and the electronic counting circuits, reference may be made to the section on "Apparatus" in their report. Suitable variations in procedures were used in the present work to search for the possible presence of protons arising from the disintegration of the deuteron.

The 14-Mev neutrons from the $T(d,n)He^4$ reaction were incident on a deuterium target located within the envelope of the counter telescope. The deuterium target was in the form of a thin layer of heavy paraffin melted in vacuum into a 0.004-inch recess in a 0.050-inch thick flat platinum button and then machined to a uniform thickness. The layer had a total weight of 15.98 mg, surface density 9.82 mg cm^{-2}, diameter about 1.5 cm, and for determination of the number of deuterium target atoms was assumed to have chemical composition $C_{45}D_{92}$. The proton impurity in the heavy paraffin was probably several percent, though there was no reliable analysis of the purity. The radiator was located inside

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[9] Gordon L. Griffith has kindly communicated to us from the University of Illinois preliminary cloud-chamber data on the angular distribution using 10 Mev neutrons. Comparison of the two sets of data shows fair agreement.
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the common envelope of the three-counter system at a distance of 18.0 cm from the neutron source.

Limiting apertures with $\frac{3}{4}$-inch diameter holes were located in front of each of the three counters. An ionizing particle ejected from the radiator would cause a double and triple coincidence count simultaneously if it passed near the axis of the counter system through all three of the limiting apertures. The distance r between the radiator and the apertures determined the solid angle for counting and was different for different sets of data as well as for double and triple coincidence. All data were adjusted, by the factor $1/r^2$, to the distance 21.0 cm which was the value applying directly to the best angular distribution data. At the distance of 21.0 cm the maximum angular range for acceptance of particles ejected from the radiator was about ± 7 degrees. To obtain angular distributions the counter assembly was mounted so as to pivot about an axis through the radiator.

PROCEDURE

In order to distinguish between recoil deuterons and disintegration protons, the detector system was operated under a variety of conditions to obtain information about the range of ejected particles. In Fig. 1 range-angle curves are plotted for the various particles which may emerge when 14 Mev neutrons are incident on the heavy paraffin radiator. These are derived from well-known range-energy curves and the energy-momentum considerations of the possible processes. Since the $n-2n$ process leads to three product particles (unless a di-neutron is ejected), the disintegration protons will presumably have a continuous range of energies. Curve c is derived by assuming the case in which two separate neutrons emerge in the same direction and with the same energy. For such a case the energy E_p of the proton is determined by

$$(E_p)^{\frac{1}{2}}=\tfrac{1}{3}(E_n)^{\frac{1}{2}}\cos\theta\pm\tfrac{1}{3}(E_n\cos^2\theta+3E_n-6Q)^{\frac{1}{2}},$$

where E_n is the energy of the incident neutron, Q is the binding energy of the deuteron, and θ is the angle in the laboratory system between the direction of the incident neutron and that of the ejected proton. If the above assumption is correct, curve c gives the upper limit of the range of disintegration protons.

Examination of the curves of Fig. 1 suggests ways of discriminating between the different possible particles, and toward this end three sets of angular distribution data were taken as indicated below. The distance from the radiator to the $\frac{3}{4}$-inch diameter limiting aperture in front of the second or third counter is denoted respectively by "doubles r" which applies to double coincidence counting, and by "triples r" which applies to triple coincidence counting.

(1) 1.8 atmos. argon in counters; doubles $r=22.12$ cm, triples $r=29.23$ cm: With this pressure a particle required 57 cm range to penetrate through the counter telescope and cause a triple coincidence count. Similarly 44 cm range was required for a double coincidence count.

(2) 0.20 atmos. argon pressure; doubles $r=14.09$ cm, triples $r=21.00$ cm: This low pressure allowed particles of relatively small range to penetrate through the counters. A range of only 4.6 cm was required for triples and of 3.2 cm for doubles.

(3) 0.20 atmos. argon pressure; a 0.007-inch platinum absorber between the second and third counters; doubles $r=13.50$ cm, triples $r=20.86$ cm: The platinum absorber thickness was chosen so as barely to stop recoil deuterons at $\theta=0°$ (100 cm range). Thus a range greater than about 110 cm was necessary for triples, whereas, as in case (2), a range of only 3.2 cm was necessary for doubles.

One further set of data was taken at 0.20 atmos. pressure after reducing the size of the collimating apertures in the counter telescope from $\frac{3}{4}$-inch diameter to $\frac{3}{8}$-inch diameter. This reduced the over-all angular spread from about $\pm 7°$ to $\pm 4°$ and was done in an attempt to determine whether the angular resolution had large effect on the shape of the angular distribution curve.

As described in reference 12, many pulses induced by neutrons and by gamma-rays in the individual counters were larger than pulses originating from the small amount of ionization of the high energy protons or deuterons. On this account considerable care had to be taken in setting the bias level on the three pulse height discriminators which eliminated small pulses. Allowing too many small pulses to pass through to the coincidence circuit would cause a high background of accidental coincidence counts. For our 1.8 atmos. data the biases were set in the same manner as described in reference 12, using plutonium 239 alpha-particles inside the counters as a reproducible calibrating standard. In the case of our 0.20 atmos. data, a proton of energy 14 Mev lost only about 35 kev by ionization in one counter. For reliable setting of biases at this low level it was found more desirable to use, as a reproducible standard of pulse height, the small

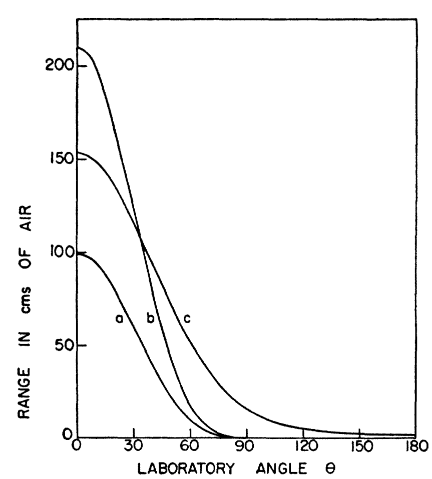

FIG. 1. Range *versus* angle for particles ejected from the heavy paraffin radiator by 14-Mev neutrons. Curve a is for recoil deuterons, curve b for recoil protons, and curve c for maximum-energy disintegration protons from $d(n,2n)p$.

TABLE I. Triple and double coincidence counts. The "Total count, rad. in" and "Total count, rad. out" correspond to the same number of monitor counts.

Recoil angle in lab system $=\theta$ Neutron angle in cm system $=\phi$	0 180	10 160	15 150	20 140	25 130	27.5 125	30 120	35 110	40 100	50 80	55 70	62.5 55	70 40	80 20
1.8 atmos. argon pressure														
Triples ($r=29.23$ cm)														
Total count, rad. in	701	200	56	52	84	37	36	22	45					
Total count, rad. out	28	6	3	9	26	13	9	9	37					
Net $\times$ $(r_1/r)^2$ per monitor unit	109	94	54	21	14	12	14	6	2					
Standard error in net	±4	±7	±8	±4	±3	±4	±4	±2	±2					
Doubles ($r=22.12$ cm)														
Total count, rad. in	1480	390	112	180	304	128	139	124	404					
Total count, rad. out	207	63	38	72	211	101	84	86	374					
Net $\times$ $(r_1/r)^2$ per monitor unit	118	91	41	30	13	8	15	11	3					
Standard error in net	±4	±6	±7	±5	±3	±4	±4	±4	±3					
0.20 atmos. argon pressure														
Triples ($r=r_1=21.00$ cm)														
Total count, rad. in	1073	934		429			383		325	466	381	189	145	124
Total count, rad. out	255	282		215			235		199	251	194	122	128	121
Net $\times$ $(r_1/r)^2$ per monitor unit	126	82		36			19		21	27	29	17	4	0.7
Standard error in net	±6	±4		±4			±3		±4	±3	±4	±5	±4	±3
Doubles ($r=14.09$ cm)														
Total count, rad. in	6216	5280		3500			4213		3298	4165	3508	1999	1879	1914
Total count, rad. out	3900	3740		3046			3871		3019	3702	3053	1750	1789	1906
Net $\times$ $(r_1/r)^2$ per monitor unit	123	87		34			19		21	26	32	28	10	0.9
Standard error in net	±5	±5		±6			±5		±6	±5	±6	±7	±7	±7
0.20 atmos. 0.007″ Pt absorber														
Triples ($r=20.86$ cm)														
Total count, rad. in	63	18		27										
Total count, rad. out	29	5		11										
Net $\times$ $(r_1/r)^2$ per monitor unit	4.2	3.2		2.7										
Standard error in net	±1.2	±1.4		±1.0										
Doubles ($r=13.50$ cm)														
Total count, rad. in	6742	2901		3920										
Total count, rad. out	4267	1928		3434										
Net $\times$ $(r_1/r)^2$ per monitor unit	128	102		33										
Standard error in net	±5	±7		±6										

gamma-ray-induced pulses from a weak radium source placed in a reproducible geometry outside the counter at the time of bias setting. The method of bias setting was as follows. Bias curves for coincidence counting of recoil deuterons at 0° (where specific ionization is a minimum) were taken, and the end of the bias plateau determined on the arbitrary but linear scale of an artificial pulse generator. To insure that high energy protons also be recorded, the operating bias was then chosen a little less than half the value at the end of the bias plateau curve, since the specific ionization of fast protons is half that of deuterons of the same energy. The counting rate with the radium source in place was then immediately determined at this bias setting and subsequently used for making small corrections in bias setting to account for daily fluctuations in gas multiplication and electronic gain.

Since the neutrons originated from the reaction $T(d,n)He^4$, the relative number of neutrons incident on the deuterium radiator in any given counting interval was monitored by counting the alpha-particles in known geometry. An alpha-particle count would enable one to determine the absolute neutron yield at any angle if either of the following two conditions existed: if the angular distribution for the $D-T$ reaction were well known, or if the alpha-particles were counted at an angle ϕ_α equal to the neutron angle ϕ_n minus 180° in the center of mass system. As to the first of these conditions, rough measurements in this laboratory indicate that the thick target neutron yield is nearly isotropic in the center of mass system for our deuteron bombarding energy of 200 kev. As to the second condition, in our geometry ϕ_n was 100°, corresponding to which the most desirable angle for counting alphas would be $\phi_\alpha=80°$. Since our alpha-counting was done at $\phi_\alpha=116°$, one would expect an angular anisotropy to have a relatively small effect on our measurement. Combining the above considerations, we estimate that anisotropy will not introduce an error of more than 5 percent in our determination of the absolute number of neutrons incident on the heavy paraffin radiator.

The coincidence counting data was taken in cycles in which a count with the heavy paraffin in place was always followed by a background count taken with the paraffin coated platinum button rotated through 180° so as to expose clean platinum in place of the heavy paraffin. Subtraction of the background count then gave the coincidence count due only to particles ejected from the heavy paraffin radiator.

RESULTS

Most of the experimental results on the angular dependence of coincidence counts are summarized in Table I. Three sets of data are given as obtained under the three sets of conditions described above. The total number of counts obtained with the radiator in place ("rad. in") and with the radiator rotated out ("rad. out") are listed and determine the statistical accuracy of the data. In analysis of the data the total count per 64,000 counts ("monitor unit") on the alpha-particle monitor is determined, and the result for "rad. out" is subtracted as a background from that for "rad. in" to determine the net coincidence count due to the presence of the radiator. The net count is then adjusted to the solid angle corresponding to the $r=21.0$ cm distance and listed in Table I under "Net $\times$ $(r_1/r)^2$ per monitor unit" along with the statistical error.

In the discussion that follows it will become evident that the net coincidence count must be almost entirely due to recoil deuterons, to the exclusion of disintegration protons or recoil protons. The first evidence for this appears in examination of the data from 0 to 20°. In this angular range the doubles and triples at 1.8 and at 0.20 atmos. all agree within statistical errors. This implies that, within our accuracy of about ± 10 percent, all particles having range greater than 3.2 cm also have range greater than 57 cm; i.e., there are no disintegration protons of energy between about 1.5 Mev and 7 Mev. Since, for the 0.20 atmos. data, there is very little difference in the range required for doubles (3.2 cm) and for triples (4.6 cm), the agreement between these data is good evidence to justify our method of adjusting the data to one solid angle by the appropriate $1/r^2$ factors.

Further evidence for the premise stated above lies in examination of the data at angles greater than 20°. With increasing angle above 20°, the 1.8 atmos. data rapidly fall below the 0.20 atmos. data. This is to be expected if the particles are recoil deuterons, since at 1.8 atmos. recoil deuterons at angles larger than 20° begin failing to have sufficient range to penetrate into the third counter if one includes the effects of radiator thickness (8.6 cm air equivalent) and angular spread. The corresponding angle for recoil contamination protons is 38°, and for maximum-range disintegration protons is 46 degrees. Similar angles applying to the occurrence of double coincidences are about 5° larger. The low value of the 1.8 atmos. datum at 40° sets an upper limit to the number of disintegration protons of energy greater than 7 Mev at this angle.

The 0.20 atmos. data, as shown in Fig. 2, form a smooth curve for angles up to about 55° where there is a fairly sudden drop, slightly more delayed for the doubles data (solid circles) than for the triples data (hollow circles). This drop must be expected for recoil deuterons, which, for angles greater than 50°, begin failing to have range enough to cause triples. The corresponding angle for recoil protons is 56° and for maximum-range disintegration protons is 86°, while the same angles applying to double coincidences are a few degrees larger. The break in the curve is not very sharp because of the effect of radiator thickness and because of lack of angular resolution. The low value of the 0.20 atmos. data in the region of 80° sets an upper limit to the number of disintegration protons near 80° of energy greater than about 1.5 Mev.

The data with the platinum absorbing foil at 0°, 10°, and 20°, also shown in Fig. 2, indicates that at 0° the number of particles of range greater than 110 cm is about 3 percent of the number of particles of range less than 110 cm. Assuming all of this 3 percent count is due to contamination protons, and using the best available values of the $n-p$ and $n-d$ differential scattering cross sections for 0° recoils, we can calculate the proton contamination in the heavy paraffin radiator. The value so obtained is 6 percent, which is not an unexpectedly high value. Thus an upper limit is set on the number of disintegration protons at 0° of range greater than 110 cm.

The above rather detailed and piecemeal evidence against the presence of disintegration protons leads the authors to believe that almost certainly the integrated cross section for the disintegration process over the range from 0° to 80° in the laboratory system and

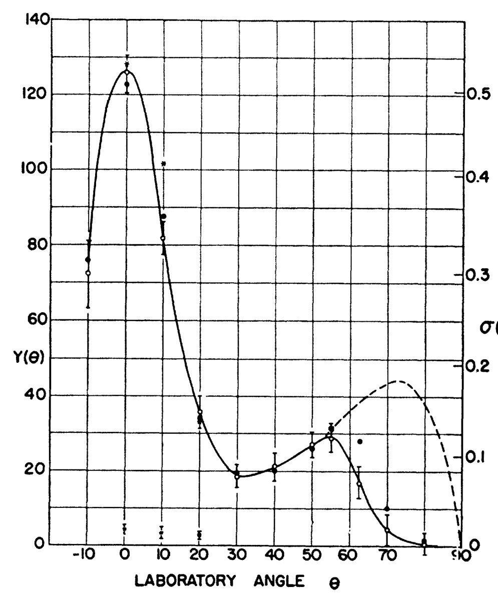

FIG. 2. Coincidence counting data as a function of laboratory angle θ for recoil deuteron. $Y(\theta)$ = net number of coincidence counts per 64,000 alpha-monitor counts. $\sigma(\theta)$ = cross section per unit solid angle. The dashed curve is an extrapolation drawn to give an area under the curve corresponding to the total scattering cross-section value taken from reference 10.

Ĭ = triples at 0.20 atmos. pressure in counters.
● = doubles at 0.20 atmos.
Ĭ = triples with 0.007″ Pt absorber, 0.20 atmos.
* = doubles with 0.007″ Pt absorber, 0.20 atmos.

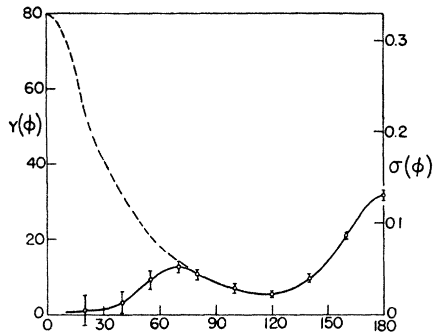

FIG. 3. Angular distribution data plotted in center of mass system. The data plotted are the triple coincidence data with 0.20 atmos. pressure in counters. The dashed curve is an extrapolation drawn to give an area under the curve corresponding to the total scattering cross-section value taken from reference 10. $\sigma(\phi)$ = scattering cross section per unit solid angle in the center of mass system.

yielding protons of energy greater than about 2 Mev is less than about 0.05 barn. It follows that the curve of Fig. 2 is essentially the $n-d$ scattering angular distribution in the range from 0 to 50°. Information about the curve in the remaining region between 50° and 90° may be gotten by extrapolation using the total cross-section values of Ageno *et al.*[7] The method of extrapolation will be described below.

The data taken with the narrow geometry for which the angular resolution was about $\pm 4°$ are compared with the $\pm 7°$ resolution data in Table II, which tabulates the ratio of the count with $\theta = 0°$ to the count with $\theta = 10°$. Within statistical accuracies the change in angular resolution has no effect on these ratios, indicating that the $\pm 7°$ resolution data are not badly distorted.

Examination of Table I shows abnormally high background coincidence rates for the 0.20 atmos. data as compared to the 1.8 atmos. data. It was shown, by measurements of the time resolution of the detecting and counting system and by recording the individual counts occurring in each of the three counters, that most of this high background was not due to accidental coincidences. It is reasonable to expect that there are many relatively short range background particles from the brass counter walls, steel diaphragms, and filling gas which can cause a high true coincidence background at 0.20 atmos. but not at 1.8 atmos.

By making observations at 0° Ageno *et al.*[7] conclude that possibly the $n-2n$ process has a cross section of the order of 0.05 barn. Our data at 0° sets an upper limit of about 0.05 barn for differential cross section in the laboratory system at 0° unless there are a relatively large number of low energy protons of energy less than 1.5 Mev. The two sets of data are therefore not inconsistent.

Figure 3 shows our angular distribution data plotted in the center of mass system, ϕ being the angle for the scattered neutron. The data plotted are the triple coincidence data at 0.20 atmos. counter pressure, and the dashed curve is the extrapolation referred to above. The extrapolation was made in two ways, and essentially the same result obtained. The first method was to assume our absolute neutron flux measurement by alpha-particle counting was correct, calculate the differential cross section at $\phi = 180°$, and draw the extrapolation with a somewhat arbitrary shape but in such a way that $\int_0^\pi \sigma(\phi)\sin\phi d\phi$ was equal to the value 0.86 barn given in reference 7. The second method used the fact that the $n-p$ scattering measurements of Barschall and Taschek had been performed with identical equipment and under directly comparable geometric conditions. The alpha-particle counter was considered not as a neutron flux measuring device but only as a flux monitor. Then the extrapolation of the $n-d$ scattering yield curve was drawn to satisfy the relation

$$\int_0^\pi Y_{nd}(\phi)\sin\phi d\phi = \frac{N_d \sigma_{nd}(\text{total})}{N_p \sigma_{np}(\text{total})} \int_0^\pi Y_{np}(\phi)\sin\phi d\phi,$$

where the $Y(\phi)$'s are relative yields per monitor count, N is the total number of deuterons or protons in the radiators, and where the $n-p$ scattering is assumed isotropic in the center of mass system. This method thus does not depend on our absolute neutron flux determination and uses the ratio $\sigma_{nd}(\text{total})/\sigma_{np}(\text{total})$ of the total cross-section values rather than either of the individual cross sections given in reference 7. Both methods determine the fraction of the area under the yield curve in the extrapolated region, but the second method does not determine the scale of values for the absolute differential scattering cross section $\sigma(\phi)$. The two methods agree as to the fraction of the area in the extrapolated region, and one therefore has more confidence in the neutron flux measurement which determines the scale of absolute values indicated in Fig. 3. The corresponding scale of values of differential cross section $\sigma(\theta)$ for the recoil deuteron yield in the laboratory system is shown in Fig. 2.

Since the total cross section is obtained by integrating the function $\sigma(\phi)\sin\phi$, our extrapolation gives a poor determination of $\sigma(\phi)$ at $\phi = 0°$. However, it is certain that there is a peak for forward scattering of the neutron which is considerably larger than the peak for backward scattering, even though the forward scattering peak is in the extrapolated region. A direct measurement of the forward scattering peak would be very desirable, but may be accessible only to measurements in which the scattered neutrons themselves are detected.

TABLE II. Effect of angular resolution.

		Doubles	Triples
$\pm 7°$ resolution:	$\frac{0° \text{ count}}{10° \text{ count}} =$	1.41 ± 0.12	1.54 ± 0.12
$\pm 4°$ resolution:	$\frac{0° \text{ count}}{10° \text{ count}} =$	1.59 ± 0.11	1.55 ± 0.22

From a comparison of our curve at 14 Mev with the theoretical curves of Buckingham and Massey[5] at 11.5 Mev, one may make the following general statements. The theoretical and the experimental curves agree in features of marked angular asymmetry with a minimum at about 110° and a forward scattering peak larger than the backward scattering peak. However, regardless of how the curves are normalized the experimental curve differs at some angles by factors of several from the various theoretical curves. As is pointed out by Massey and Buckingham,[4] their "exchange force" calculations will probably come into better agreement with experimental *total* cross-section values[7] in the region of 14 Mev when they take into account the contributions of higher than p angular momentum states, which were ignored in their original calculations. Such a refinement in the "exchange force" theory may also bring it into better agreement with our angular distribution data. If such an extension of the theory gives favorable results, the *over-all* evidence will be somewhat more in favor of "exchange forces" than "ordinary forces." However, as Darby and Swan[8] point out, their angular distribution data at 2.5 Mev agrees very closely with the "ordinary force" curve of Buckingham and Massey, whereas the data of Coon and Barschall[6] at this energy fits better with the "exchange force" curve. Obviously more accurate and complete data are necessary.

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Some Properties of Superconductors below 1°K. I. Titanium

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Magnetic measurements on titanium metal of purity 99.95 percent have been carried out down to 0.3°K. The titanium was found to be superconductive with a transition temperature of 0.53°K. Measurements of the magnetic threshold curve were made for which the initial slope was found to be 470 gauss per degree.

INTRODUCTION

IN carrying out investigations on techniques required for the observation of nuclear paramagnetism, the properties of some superconductors have been studied below 1°K. Details of the results obtained with high purity titanium are given herewith.

Titanium of stated purity approximately 99.75 percent was found by Meissner[1] in 1930 to become superconductive by observation of the resistance of a single crystal specimen. The transition point in zero magnetic field was found to be 1.13°K. Subsequent resistance measurements by Meissner, Franz, and Westerhoff[2] on other titanium samples showed superconductive transitions at higher temperatures, one being as high as 1.77°K. de Haas and van Alphen[3] using titanium of unstated purity found by resistance measurements a superconductive transition at about 1.72°K, with a broad transition region. Webber and Reynolds[4] in 1948 observed an anomaly in the resistance of a titanium specimen, prepared in a different manner from the above, the resistance showing a marked decrease over a broad temperature range between 3°K and 1°K without, however, showing superconductivity.

Measurements by Shoenberg,[5] on the other hand, on the magnetic properties of titanium of purity 99.9 percent revealed no sign of superconductivity, defined by the establishment of zero magnetic induction throughout the entire specimen, down to temperatures of about 1.0°K. Shoenberg[5] reported, however, the occurrence of a very small diamagnetic anomaly at about 1.5°K and suggested that the previous observations of zero resistance at about this temperature were due to very small regions in the specimen becoming superconductive where chemical or physical impurity might have been concentrated. Such a view is supported by the considerable scatter in the values of transition points observed.[1–3]

The experiments reported in this paper were undertaken in order to clarify the question of the possible occurrence of superconductivity in titanium which in view of Shoenberg's work appeared doubtful. From the position of titanium in the periodic table it would appear that it should become superconductive, since the other metals in the same sub-group are super-

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