The Differential and Total Neutron Scattering Cross Sections of the Deuteron in the Energy Range 0.1 to 1.0 Mev

P. R. TUNNICLIFFE*
Atomic Energy Research Establishment, Harwell, Berkshire, England
(Received December 8, 1952)

The angular distribution of neutrons scattered by deuterons has been investigated by proportional counter techniques in the energy range 0.1 to 1.0 Mev. The distribution shows a very marked anisotropy at the higher energies which is still appreciable at 135 kev. Values for the total scattering cross sections are in agreement with earlier data down to 446 kev but apparently increase above the accepted value for the unbound thermal cross section.

I. INTRODUCTION

In view of the large dimensions of the deuteron, it is expected that neutron scattering experiments at relatively low neutron energies will yield information concerning the nature of nuclear forces. Theoretical treatment of this three-body problem has been carried out by Buckingham and Massey,[1] and by Verde,[2] and recently by Buckingham, Hubbard, and Massey.[3] The treatment essentially yields isotropic scattering at neutron energies below 1 Mev.

Most of the experimental work published relating to the differential scattering cross section has been carried out for neutrons of about 2.5-Mev energy (Coon and Barschall;[4] Kruger, Shoupp, Watson, and Stallman;[5] Darby and Swann;[6] Caplehorn and Rundle[7]). The experimental results are somewhat conflicting, as pointed out by Caplehorn and Rundle. The later results favor ordinary or neutral type nuclear forces, while the earlier results are in better agreement with exchange or symmetrical theory forces. Recent work by Hamonda and de Montmollin[8] and by Wantuch[9] at higher neutron energies suggests that the theory is not adequate. The total scattering cross-section measurements of Aoki,[10] Nuckolls *et al.*,[11] Rainwater *et al.*,[12] Fermi and Marshall,[13] and Wollan, Schull, and Koehler[14] strongly favor the exchange type of force. Recent measurements of both the differential and total scattering cross sections have been reported by Walt, Okazaki, and Adair.[15] Their results support those of Coon and Barschall.

The corresponding theoretical treatment of neutron-proton scattering does not expect any appreciable anisotropy in the scattering at neutron energies of less than 10 Mev, and this has been confirmed by a number of authors (i.e., Kruger, Shoupp, and Stallman,[16] Bonner,[17] Coon and Barschall,[4] Laughlin and Kruger,[18] Barschall and Taschek,[19] Caplehorn and Rundle[7]). This isotropic scattering has been used as a cross check of the present experimental work.

II. EXPERIMENTAL METHOD

The measurements reported in this paper are based on a method described by Barschall and Kammer.[20] The pulse size distribution from a proportional counter containing deuterium gas is observed when the counter is irradiated with monoenergetic neutrons. The pulse size distribution is a direct measure of the distribution in energy of the recoiling deuterons, except for corrections for wall and end effects in the counters. We assume that, in the scattering of neutrons by deuterons in the energy range reported, only S and P wave scattering terms need be taken into account. Parallel observations were taken with a hydrogen filled counter to ensure that the method yielded isotropic scattering by protons.

The method of interpretation of the pulse distributions is outlined below. We use the notation E_n = neutron energy, E = recoil energy, E_m = maximum recoil energy, ϕ = angle of recoiling nucleus (laboratory system), θ = angle of scattering (center-of-mass system), $\sigma(\theta)$ = differential scattering cross section (center-of-mass system), σ_s = total scattering cross section, and M = mass of recoiling nucleus relative to the neutron mass. Then, from the conservation of momentum, E_m

* Now with Atomic Energy of Canada, Limited, Chalk River, Ontario, Canada.
[1] R. A. Buckingham and H. S. W. Massey, Proc. Roy. Soc. (London) **A179**, 123 (1941); Phys. Rev. **71**, 558 (1948).
[2] M. Verde, Helv. Phys. Acta **22**, 339 (1949).
[3] Buckingham, Hubbard, and Massey, Proc. Roy. Soc. (London) **A211**, 183 (1952).
[4] J. H. Coon and H. H. Barschall, Phys. Rev. **70**, 592 (1946).
[5] Kruger, Shoupp, Watson, and Stallman, Phys. Rev. **53**, 1014 (1938).
[6] J. F. Darby and J. B. Swan, Nature **161**, 22 (1948).
[7] W. F. Caplehorn and G. P. Rundle, Proc. Phys. Soc. (London) **64**, 546 (1951).
[8] I. Hamonda and G. de Montmollin, Phys. Rev. **83**, 1277 (1951).
[9] E. Wantuch, Phys. Rev. **84**, 169 (1951).
[10] H. Aoki, Proc. Phys.-Math. Soc. Japan **21**, 75 (1939).
[11] Nuckolls, Bailey, Bennett, Bergstralh, Richards, and Williams, Phys. Rev. **70**, 805 (1946).
[12] Rainwater, Havens, Dunning, and Wu, Phys. Rev. **73**, 733 (1948).
[13] E. Fermi and L. Marshall, Phys. Rev. **75**, 578 (1949).
[14] Wollan, Schull, and Koehler, Phys. Rev. **83**, 700 (1951).
[15] Walt, Okazaki, and Adair, Phys. Rev. **87**, 238 (1952).
[16] Kruger, Shoupp, and Stallman, Phys. Rev. **52**, 678 (1937).
[17] T. W. Bonner, Phys. Rev. **52**, 685 (1937).
[18] J. S. Laughlin and P. G. Kruger, Phys. Rev. **73**, 197 (1948).
[19] H. H. Barschall and R. F. Taschek, Phys. Rev. **75**, 1819 (1949).
[20] H. H. Barschall and M. H. Kammer, Phys. Rev. **58**, 590 (1940).

$=4ME_n/(M+1)^2$ and $E=E_m\cos^2\phi$. Since $\theta+2\phi=\pi$,

$$E=E_m(1-\cos\theta)/2. \quad (1)$$

If $p(E)$ is the probability per unit energy interval of a recoiling nucleus attaining energy E,

$$p(E)=\sigma(\theta)\pi(M+1)^2/\sigma_s E_n M. \quad (2)$$

Relations (1) and (2) show that the pulse size distribution is equivalent to a measurement of the differential scattering cross section in terms of $\cos\theta$.

In the case of hydrogen $\sigma(\theta)/\sigma_s=1/4\pi=$ constant, and we obtain a uniform distribution.

The proportional counters used in these measurements have a cylindrical sensitive volume whose ends are electrically well defined and have been described in detail by Skyrme, Tunnicliffe, and Ward.[21] When neutrons are directed axially, the method of allowing for wall and end effects (i.e., for recoil particles leaving or entering the ends or striking the walls before expending all their energy) has been detailed for hydrogen. The pulse distribution for hydrogen is of the form

$$\phi_H(x)=1+\frac{R_0}{a}\frac{dI_1}{dx}+\frac{R_0}{b}\frac{dI_2}{dx}-\frac{R_0^2}{ab}\frac{dI_3}{dx},$$

where $\phi_H(x)$ is proportional to the number of pulses of size x per unit interval of size, $R_0=$ maximum recoil proton range, $a=$ radius, and $b=$ length of sensitive volume of the counter, and where the normalization is such that $x=1$ is the maximum pulse size. The correction functions I_1, I_2, and I_3 have been computed.[21]

If we assume that the differential cross section of the deuteron can be written

$$\sigma(\theta)=\sigma_0(C_1+C_2\cos\theta+C_3\cos^2\theta)/4\pi C_1, \quad (3)$$

it can be shown that the pulse distribution from the deuterium-filled counter is of the form

$$\phi_D(x)=C_1\phi_1(x)+C_2\phi_2(x)+C_3\phi_3(x), \quad (4)$$

where

$$\phi_1(x)=1+\frac{R_0}{a}\frac{dI_4}{dx}+\frac{R_0}{b}\frac{dI_5}{dx}-\frac{R_0^2}{ab}\frac{dI_6}{dx},$$

$$\phi_2(x)=2(1-2x)+\frac{R_0}{a}\frac{dI_7}{dx}+\frac{R_0}{b}\frac{dI_8}{dx}-\frac{R_0^2}{ab}\frac{dI_9}{dx},$$

$$\phi_3(x)=(1-2x)^2+\frac{R_0}{a}\frac{dI_{10}}{dx}+\frac{R_0}{b}\frac{dI_{11}}{dx}-\frac{R_0^2}{ab}\frac{dI_{12}}{dx}.$$

The functions I_4, $I_5\cdots I_{12}$ arise from end and wall effects. They have been computed by methods similar to those outlined earlier[21] but assuming a linear range-energy relation for deuterons in the energy range of interest. This assumption should not give rise to inaccuracies in these functions of more than a few percent and has been a considerable saving in computing effort.

[21] Skyrme, Tunnicliffe, and Ward, Rev. Sci. Instr. **23**, 204 (1952).

The observed pulse distributions for deuterium were interpreted as follows. The best values of the coefficients C_1, C_2, C_3, and their standard deviations were obtained by making a least squares fit of Eq. (4) to the observations. From measurements with the hydrogen counter a value Φ was obtained for the total number of neutrons incident on the sensitive volume of the deuterium counter during the time taken to observe the pulse distribution. The value of σ_0 was obtained using the relation $C_1=\Phi b\rho\sigma_0$, which holds for the normalizations used, where $b=$ length of the sensitive volume of the counter and $\rho=$ number of deuterium atoms/cc of the filling. Integration of Eq. (3) gives the total scattering cross section,

$$\sigma_s=\sigma_0(1+C_3/3C_1). \quad (5)$$

Finally Eq. (3) was written in the more conventional form,

$$\sigma(\theta)=\sigma_0(1+\alpha\cos\theta+\beta\cos^2\theta)/4\pi, \quad (6)$$

where $\alpha=C_2/C_1$ and $\beta=C_3/C_1$.

III. EXPERIMENTAL APPARATUS

Monoenergetic neutrons were obtained from the $Li^7(p,n)Be^7$ reaction. An analyzed proton beam of energy up to 2.5 Mev from the Harwell electrostatic generator[22] with beam currents of 4μa was used. Targets of evaporated lithium fluoride on silver backings of thickness between 5 and 30 kev were mounted at the end of a thin aluminum thimble six inches long. The energy spread of the proton beam was controlled to about ±1 kev at the lower energies by an electron gun stabilizer and to about ±5 kev at higher energies by a slit placed in front of the target. Machine voltages were measured with a generating voltmeter calibrated using the $Li^7(p,n)Be^7$ threshold and neutron energies calculated using the known proton voltage.

Two counters were suspended in a light aluminum framework in a vertical plane intersecting the target and containing the proton beam and were orientated at angles of 20° above and below the direction of the beam. The counters were each about two and a half feet from the target. Apart from the beam analyzing magnet the nearest substantial mass of material was about nine feet from the target and counters.

One counter was filled with either pure deuterium at 78.1-cm pressure of mercury (16.3°C) or 30.7 cm of deuterium, 50.1 cm of argon, and 1.8 cm of carbon dioxide (20°C), and the other with either 79.6 cm of pure hydrogen (16.3°C) or 78.0 cm of pure methane (20°C). The fillings mentioned first were used for measurements below 450-kev neutron energy. The pure methane, hydrogen, and carbon dioxide were prepared by fractional distillation.[23] The deuterium was prepared by

[22] R. L. Fortescue and P. D. Hall, Proc. Inst. Elec. Engrs. (London) **96**, 77 (1949).

[23] The author is indebted to Dr. London of the Atomic Energy Research Establishment for preparation of the pure methane and to the Clarendon Laboratory, Oxford for the pure hydrogen.

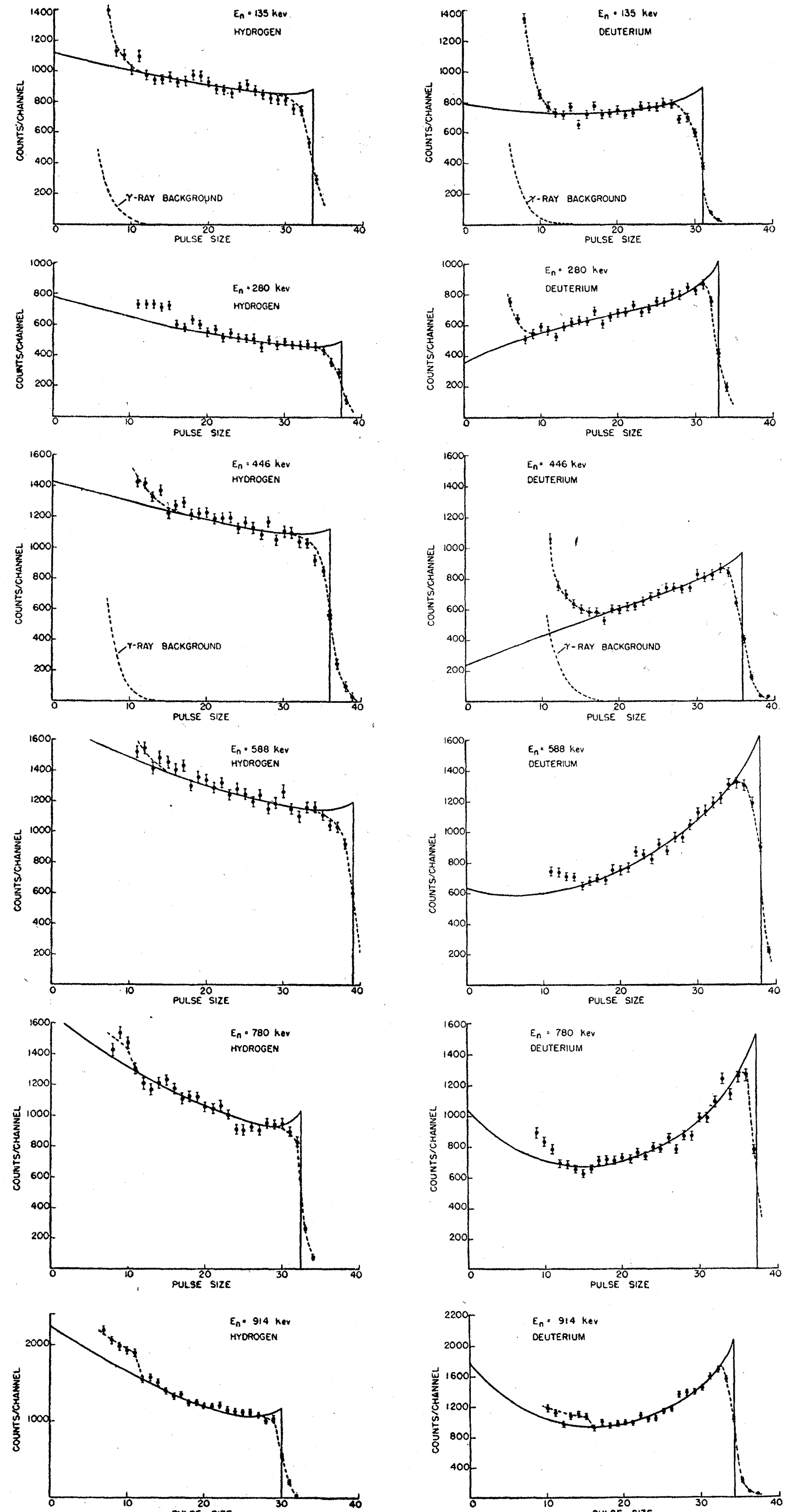

FIG. 1. Comparative pulse distributions obtained at various neutron energies E_n from counters containing hydrogen and deuterium. The solid lines are calculated assuming isotropic scattering in hydrogen and a mixture of S and P wave scattering in deuterium, allowing for end and wall effects.

TABLE I. Experimental values of the constants of Eqs. (5) and (6).

E_n kev	α	β	σ_0 cm$^2\times10^{-24}$	σ_s barns
135	-0.099 ± 0.039	0.111 ± 0.146	$4.6\ \pm0.07$	$4.8\ \pm0.34$
280	-0.470 ± 0.022	0.075 ± 0.082	3.70 ± 0.06	3.79 ± 0.22
446	-0.419 ± 0.092	0.055 ± 0.224	3.04 ± 0.09	3.10 ± 0.29
588	-0.496 ± 0.014	0.602 ± 0.125	2.62 ± 0.04	3.14 ± 0.20
780	-0.467 ± 0.036	0.838 ± 0.105	2.44 ± 0.04	3.18 ± 0.17
914	-0.333 ± 0.052	$1.17\ \pm0.13$	2.37 ± 0.04	3.29 ± 0.20

electrolysis of heavy water and purified by the method described by Wilson *et al.*[24] Mass spectrometer analyses[25] established the isotopic purity to be 98.25 percent D_2, 1.75 percent H_2.

Pulses from the counters were amplified by A.E.R.E. type 1008 linear amplifiers using the high frequency head amplifiers and fed in turn to a 30-channel pulse analyzer. Runs with each counter at a given neutron energy were taken with and without a neutron 'shadow cone' inserted between the counter and target to allow a correction to be made for scattered neutrons. This shadow cone consisted of a thin-walled aluminum can six inches long filled with water. Runs were monitored by measuring the integrated proton current received by the target.

The pulse distributions from the hydrogen containing counter were fitted to calculated curves immediately after they were obtained and showed that satisfactory experimental conditions had been established and were maintained during the series of measurements. They were used to determine the number of neutrons incident on the counter and hence on the deuterium counter during the corresponding run with the latter, after allowing for a small inverse square correction and using the integrated proton currents received by the target during the runs.

Runs were taken below the neutron threshold of the $Li^7(p, n)Be^7$ reaction with each filling to assess the gamma-ray background and to assist in rejecting points at the lower end of the distributions which might be modified by this background.

IV. RESULTS

The pulse distribution of comparison runs for the hydrogen and deuterium-filled counters are shown in Fig. 1. These data have been corrected for variations in pulse analyzer channel widths, for hydrogen content of the deuterium counter, and for scattered neutron background. The errors shown are standard deviations of the number of counts per channel. The solid lines are the theoretical curves calculated by the methods outlined in Sec. II. The maximum pulse size used in adjusting the pulse size scale of Eq. (4), and therefore entering into the value of C_1, has been assessed by taking the value at which the actual distribution falls to half the extrapolated value of the distribution which might be expected but for resolution effects. From previous experience[26] this assessment should not be in error by greater than 1 percent.

Correction for the hydrogen recoils in the deuterium counter was obtained in a straightforward manner using the hydrogen counter distribution at the corresponding neutron energy and allowing for the ratio of hydrogen in two counters and the lengths of the runs. Over the range of pulse sizes used, the scattered neutron background never amounted to more than a few percent. In carrying out the least squares fit of Eq. (4) to the experimental results the distributions were inspected and points at the upper and lower end of the runs discarded before making the analysis. In general, it was necessary to discard the upper three or four points since the upper ends of the distributions were smeared out by resolution effects in the counter and by the finite width of the incident neutron spectrum. The lower ends of the distributions were discarded where contributions of more than a few percent might have been made by the gamma-ray background and where a contribution could arise from the second neutron group from the $Li^7(p, n)Be^7$ reaction.[27]

The values of the constants α, β, σ_0, and σ_s obtained are listed as a function of neutron energy in Table I. The errors quoted for α, β, and σ_0 are standard deviations obtained from the analysis using the statistical accuracy of the experimental data. The restricted angular range has been allowed for automatically. A further uncertainty of 5 percent, assessed somewhat arbitrarily, has been added to the error of σ_s to allow for uncertainties in the neutron flux determination.

In treatment of the results it has been assumed that the ionization potential for the low energy recoil protons and deuterons used in this work is constant within the experimental accuracy and, therefore, that the pulse sizes were proportional to the recoil energies. This assumption is based on the work of Tunnicliffe and Ward.[26]

V. DISCUSSION AND CONCLUSIONS

It is concluded that neutron-deuteron scattering is markedly anisotropic at neutron energies of about 1 Mev. The anisotropy decreases with neutron energy but is still appreciable at 135-kev neutron energy. This magnitude of the anisotropy does not appear to be predicted by existing theoretical treatments. However, Buckingham, Hubbard, and Massey[3] point out that their calculations cannot be relied upon for neutron energies less than 2 Mev and propose to extend their calculations in more detail at low neutron energies. If an extrapolation of the trend of the experimentally observed anisotropy is permissible, it may be concluded

[24] Wilson, Beghian, Collie, Halban, and Bishop, Rev. Sci. Instr. **21**, 699 (1950).

[25] The author is indebted to the mass spectrometer group at the Atomic Energy Research Establishment for these analyses.

[26] P. R. Tunnicliffe and A. G. Ward, Proc. Phys. Soc. (London) **65**, 233 (1952).

[27] B. Hammermesh and V. Hummel, Phys. Rev. **78**, 73 (1950).

that these results tend to support those of Darby and Swan[6] and of Caplehorn and Rundle.[7]

The total cross-section values are in agreement within the experimental errors with the results of Nuckolls *et al.*[11] down to the lowest energies which they used (350 kev). The value 3.79 barns at 280 kev agrees within the experimental errors with the results of Walt, Okazaki, and Adair.[15] Both this value and the value of 4.8 barns at 135 kev are high in comparison with the unbound values of 3.3, 3.44, and 3.4 barns at very low neutron energies given, respectively, by Rainwater *et al.*,[12] Fermi and Marshall,[13] and by Wollan, Schull, and Koehler.[14] Owing to pressure of circumstances, it was not possible to repeat measurements and obtain adequate cross checks of these apparently anomalous cross sections in the time available. It would therefore seem desirable that the total cross section be investigated further below 500 kev.

The author is indebted to Dr. T. H. R. Skyrme for development of the method of handling the wall corrections, to Miss P. Dyson for much of the extensive numerical work, and to Mr. A. E. Pyrah for efficient maintenance and operation of the electrostatic generator. He wishes to thank the Director of the Atomic Energy Research Establishment for permission to publish this paper.

PHYSICAL REVIEW VOLUME 89, NUMBER 6 MARCH 15, 1953

A New Rigorous Lower Bound on the Range of the Triplet Neutron-Proton Interaction*

LESLIE L. FOLDY
Case Institute of Technology, Cleveland, Ohio
(Received December 1, 1952)

The experimental values of the electric quadrupole moment and percentage D state in the deuteron together with the effective triplet range of the neutron-proton interaction are employed to determine a rigorous lower bound on the "cut-off" range of the neutron-proton interaction. The latter is defined to be the range beyond which the deuteron wave functions have effectively their asymptotic form.

It was first pointed out by Schwinger[1] that the existence of an electric quadrupole moment for the deuteron requires that the neutron-proton interaction in the triplet state have a finite range. By the use of approximate expressions for the deuteron quadrupole moment and percentage D state in the deuteron, Schwinger was further able to make an estimate of a lower bound on this range of 2.5×10^{-13} cm. More recently Broyles and Kivel[2] introduced an ingenious method for obtaining a *rigorous* lower bound on the triplet range. Before describing their approach it is necessary to give a more precise definition of the term "range" as it is used above. It is used here in the sense of the neutron-proton separation beyond which the deuteron S and D wave functions have effectively their asymptotic form. For an interaction which does not have a tail but cuts off sharply, it then represents the separation at which the cutoff occurs. For a long-tailed interaction, a precise definition is not so easy to formulate, and one must be satisfied with defining it as the neutron-proton separation beyond which the deviation of the actual deuteron wave function from its asymptotic form is sufficiently small to make no difference within the accuracy desired in the calculation of such quantities as the quadrupole moment, percentage D state, effective range, etc. To distinguish this range from such quantities as the effective range and intrinsic range, we shall refer to the range here defined as the "cut-off" range of the potential.

The method of Broyles and Kivel consists in completely ignoring the form of the interaction in the deuteron and concentrating attention on the deuteron wave function. Assuming then that these wave functions have their asymptotic form outside the cut-off range of the potential and an arbitrary form inside, they maximize the expression for the quadrupole moment of the deuteron by variation of the form of the wave functions inside the range of the interaction and the amplitude of the asymptotic forms outside the range of the interaction. This maximum quadrupole moment is then a monotonically increasing function of the range of the interaction, and the observed quadrupole moment then sets a lower bound on the range of the interaction. It is worth noting that the maximum quadrupole moment for a fixed range is obtained by having the wave functions vanish inside the range of interaction. The lower bound which these authors find for the cut-off range turns out to be 1.1×10^{-13} cm which is considerably below that estimated by Schwinger.

At least part of this last discrepancy can be at-

* This work was supported by the U. S. Atomic Energy Commission. Part of the work herein contained was carried out while the author was a summer visitor at Brookhaven National Laboratory. The hospitality extended to the author by this laboratory is gratefully acknowledged.
[1] J. Schwinger, Phys. Rev. **60**, 164 (1941).
[2] A. A. Broyles and B. Kivel, Phys. Rev. **77**, 839 (1950).