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The Capture Cross Section of Thermal Neutrons in Water

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Abstract. The capture cross section of slow neutrons by protons has been measured by following the decay of neutrons from a pulsed source in water.

The decay was measured by detecting the capture γ-rays with a liquid scintillator.

The observed average life in water was $203{\cdot}3 \pm 2{\cdot}6\,\mu$sec corresponding to a capture cross section of $0{\cdot}335 \pm 0{\cdot}004$ barns at a velocity of 2200 metres/sec.

§ 1. Introduction

The cross section for the photo disintegration of the deuteron, d(γ, n)p near the threshold is an important constant in nuclear theory. An exact value is of particular interest because, near threshold, the disintegration is entirely due to the photomagnetic transition in which the angular momentum is conserved by spin flipping; it also provides the most direct measure of the difference between the singlet and triplet effective ranges. A full account of the theoretical aspects of the problem has been given by Squires (1952). A direct measurement near threshold of the photo disintegration process is not possible, but as was early pointed out by Bethe (1947) the cross section σ_{cap} of the inverse capture process can be measured directly, and is related by the theory of detailed balancing to the cross section σ_{mag} for disintegration by a quantum of energy $\hbar\omega$ by the following relationship.

$$\frac{\sigma_{\text{cap}}}{\sigma_{\text{mag}}} = 6\left(\frac{\hbar\omega}{mcv}\right)^2.$$

The capture cross section varies inversely as the relative velocity v, so that, if neutrons are released in an infinite hydrogenous medium, the total number present will decrease exponentially with time, by capture, and a measurement of the average life $T = 1/\lambda$ will enable the capture cross section to be measured in terms of the composition of the medium. The capture cross section of oxygen (U.S.A.E.C. 1952) is so small that water is a very convenient hydrogenous medium to employ.

Two methods have been used to measure the average life of neutrons in a water tank:

(*a*) The measurement of the total number of neutrons N in the water tank at a single instant of time, when they are introduced at a steady rate n neutrons per second. Clearly the average life $1/\lambda = n/N$. Owing to the difficulty of making absolute neutron measurements the method cannot be used to make absolute measurements of T. It can, however, be used to give a comparison of the capture cross section of neutrons by protons and of neutrons by another nucleus of large capture cross section which also obeys the $1/v$ law, e.g. boron.

As was first pointed out by Whitehouse (Thode *et al.* 1948) the isotopic composition of the particular boron sample must also be measured. Apart from the disadvantage of not giving an absolute value, the static method runs into other difficulties and after careful consideration we decided that we were unlikely to be able to improve on the measurements of Whitehouse and Graham (1947). However, if the initial constant source of neutrons has no fast neutrons in it which have to be slowed down, these difficulties are more easily overcome, and measurements have been made (Hamermesh, Ringo and Wexler 1953) using thermal neutrons from the Argonne pile. Although precise, their results are tied to the still uncertain capture cross section of natural boron (Green *et al.* 1954). (*b*) Direct measurement of the decay from a pulsed neutron source. This was first tried by Manley, Haworth, and Luebke (1941, 1942) and more recently by Von Dardel and Waltner (1953) and Von Dardel and Sjostrand (1954). The dynamic method suffers from several fundamental difficulties: Firstly, the motion of the neutrons during their life must be allowed for since the local neutron concentration changes both by capture and diffusion. Secondly, if neutron detectors are used to follow the decay it is not easy to arrange that they actually measure what is required, namely the concentration of neutrons which would have been present at the position occupied by the detector had the medium not been disturbed by the presence of the detector. Thirdly, it is difficult to obtain a sufficient neutron flux to obtain good statistical accuracy.

Since it seems impossible to make the measurements without having to make corrections, we attempted to ensure that any correction should not exceed 10% of the quantity measured and could be measured experimentally in a subsidiary experiment to an accuracy of at least 10%. Working in this way, we did not have to rely entirely upon theoretical calculations in applying the correction, and hoped to arrive at a result which could be relied upon to $\pm 1\%$.

§ 2. The Experimental Method

An outline of the apparatus used in the final measurements is shown in figure 1. The source of neutrons is a heavy ice target on which falls a pulsed beam of 450 kev deuterons. In spite of the complication of liquid air cooling,

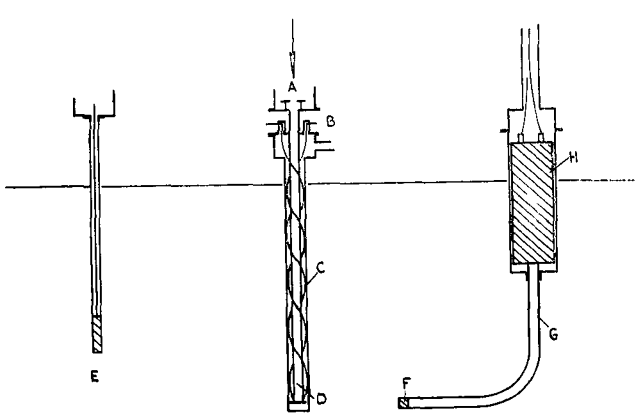

Figure 1. Heavy ice target and detector. A. Diaphragm to define the deuteron beam; B. liquid air inlet; C. vacuum jacket; D. heavy ice target; E. BF_3 neutron monitor; F. scintillator; G. Perspex light guide; H. photomultiplier.

the heavy ice target was used to get enough neutrons in each pulse (10^5 neutrons per pulse). Preliminary experiments showed that a simpler occluded target did not yield enough neutrons. While equally good or better neutron yields could be obtained with a DT source using a zirconium tritide target the large range of 14 MeV neutrons in water prevented the use of this otherwise very convenient source. Using the DD source it was possible to use a 600 gallon tank, whose dimensions were 120 cm × 120 cm × 180 cm. The neutron decay was followed by detecting the 2·2 MeV capture γ-rays with a liquid scintillator (terphenyl in cyclohexane with diphenyl hexatriene as frequency shifter) mounted on the end of a long light guide. The photomultiplier was more than 40 cm from the scintillator and the preamplifier was outside the water tank altogether. This method of detection has several advantages. It is sensitive; the cross section of the scintillator and light guide both for neutron capture and neutron scattering is so close to that of water that the disturbance of the neutron field due to the detector is negligible. Moreover, owing to the large range of γ-rays in water ($\sim$25 cm) the detector effectively integrates over a large volume, so that the effect of any small disturbance in the neutron density due to the detector is greatly reduced. This behaviour is in marked contrast to that of a conventional BF_3 neutron detector which can only with difficulty be adjusted to have the same capture cross section as that of the water it displaces, and must always have a quite different scattering cross section.

The main disadvantage of this type of detector is that it does not give the same pulse height for each γ-ray detected, and so its sensitivity depends critically on the stability of the associated electronic equipment.

This difficulty was overcome in two ways. All equipment (amplifier type 201 H.T. to photomultiplier type 1007C and scaler type 1009B) was electronically stabilized and fed from a stabilized ($\pm$ 1 volt) mains supply. Measurement of the changes of sensitivity over long periods showed that the stability was just sufficient for our purpose. Small fluctuations about the mean sensitivity do not affect the measured lifetime; any long term drift could be detected and allowed for by suitable design in the delay sorter and in fact no errors due to electronic instability could be detected.

§ 3. The Delay Sorter

The neutron decay was followed by counting the γ-rays produced from an initial 100 μsec pulse of neutrons, in successive 50 μsec intervals, the whole cycle being repeated every 1600 μsec. A series of accurately spaced pulses controlled the following series of events. The radio-frequency oscillator running the ion source switched on for 100 μsec giving a pulse of neutrons from the heavy ice target. The subsequent γ-ray pulses produced by the capture of this group of neutrons were counted by a group of four gated scalers. The gates were arranged so that counts were made in the three intervals 200–400 μsec (group B) 400–600 μsec (group C) 600–800 μsec (group D) as determined by a manually controlled selector switch.

The individual scalers were gated so as to record pulses in four successive 50 μsec channels. It would have been possible to count in all twelve channels using twelve scalers, but these were not easily available and it was found quite satisfactory to follow the decay in successive 200 μsec periods.

The gating pulses were obtained from a 20 kc/s blocking oscillator feeding two scale of four ring circuits. Accuracy of timing was ensured by locking the oscillator on to a 100 kc/s crystal controlled master oscillator. Owing to the use of double triodes (CV858) the gating pulses had a 1 μsec rise time. This was allowed for by calibrating the channels with random pulses from a radioactive source. This calibration was made frequently during the course of the measurements. A very small correction was made for differences in the decay in the channels of unequal length.

§ 4. The Ion Source

A conventional radio-frequency ion source (Thonemann 1948) was driven from a pulsed radio-frequency supply; in order to obtain regular striking the instantaneous radio-frequency power input was raised to 1·5 kilowatts. As the duty cycle was only 1/16, the mean dissipation was less than the usual 200–300 watts and no troubles due to over-running were experienced. However, the large wall absorption at the beginning of each pulse sufficiently reduced the instantaneous pressure to cause unsteady running. This trouble was overcome by replacing the dome of the discharge tube with a spherical volume of from 1 to 2 litres capacity.†

The maximum beam current used was about 1 mA corresponding to about 70 μA mean current. The liquid air cooling was just adequate to maintain the heavy ice target under this heavy bombardment. Since the slowing down time of the neutrons in the water is only a few microseconds, and no measurements were made for 100 μsec, it is not very important that the beam should end sharply. It is, however, important that there should be no neutron production during the time the decay is being measured. This was verified by displaying the amplified beam current on a cathode-ray tube. Incipient deterioration of the discharge conditions was readily detected by the appearance of large current spikes in the dead time which disappeared when the conditions in the radio-frequency arc were changed. It is clear that any failure in this respect would increase the measured value of the half-life of the neutrons.

§ 5. Measurement of Decay Time

When all the equipment was working satisfactorily the actual measurement of the neutron decay presented little difficulty. The neutrons were monitored by a fixed BF_3 counter placed in the tank 25 cm from the target and diametrically opposite the scintillator. A separate scaler was also used to count all the γ-ray pulses from the scintillator which entered channel group D(600–800 μsec) irrespective of which group was being operated. The background correction offered some difficulty since the metal cell containing the scintillator liquid became slightly radioactive owing to the neutron bombardment. The original cell was made from aluminium, but a change to a stainless steel cell very much reduced this unwelcome effect. The residual error due to this cause was eliminated by investigating the rise and fall of the activity empirically at various positions in the tank. A correction curve was then constructed based on a particular schedule of measurements which was carefully adhered to.

† We have to thank Dr. Thonemann for suggesting this remedy

At the beginning of each run measurements were first made in channel group D, so that the smallest γ-ray count also had the smallest correction for neutron induced activity.

Correction for induced activity was only necessary when the scintillator was less than 40 cm from the target.

§ 6. Normalization of Counts

All measurements were normalized to a constant number of counts in the BF_3 chamber used as a neutron monitor. This was frequently calibrated with a 20 mc Ra–Be source but no significant changes in its sensitivity were found. Small changes in the gain of the amplifier do not affect the decay time observed in one group of channels but must be avoided if the measurements in the three groups of channels at one place in the tank are to be combined into a single decay curve covering the whole 600 μsec decay period. This was done by measuring the total counts in group D independently. Only those runs were accepted in which the group D counts bore a constant ratio to the neutrons counted by the monitor, thus showing that the amplifier gain had not changed.

§ 7. Correction for the Target Tube

The tube through which the deuterons reach the target is an unavoidable part of the apparatus, whose presence must be allowed for. It affects the observed results in two ways.

Firstly, owing to its presence the initial neutron distribution is less symmetrical than the ideal distribution which would have been obtained from a source entirely surrounded by water.

Secondly, some slow neutrons will leave the tank during the period of measurement by diffusion up the target tube which would otherwise have been captured and contributed to the γ-ray count. The effect of the target tube was estimated experimentally by introducing a dummy vertically below it. The thickness of the walls of the dummy was chosen so as to have the same capture cross section for slow neutrons as that of the actual target tube. Measurements with and without the dummy tube in position enabled an estimate founded on experiment to be made of the effect of the target tube on the results.

§ 8. Experimental Results

Four independent sets of measurements were made, namely: measurements with the γ-ray detector along the horizontal and vertical axes through the target; measurements with a BF_3 neutron detector along the horizontal and vertical axes.

The following subsidiary measurements were also made with the object of placing the corrections upon a sound experimental basis: measurements with the γ-ray detector along the horizontal axis in the presence of the dummy target; measurements with the BF_3 chamber of the angular distribution of the neutron density about the target.

The variations of the neutron density in time and space are shown in figure 2. The outward diffusion of the neutrons during the capture process is clearly shown in these curves by the outward migration of the maxima. The difference between the measurements along the horizontal and vertical axis is nearly all due to the asymmetry in the angular distribution of the neutrons from the D-D reaction.

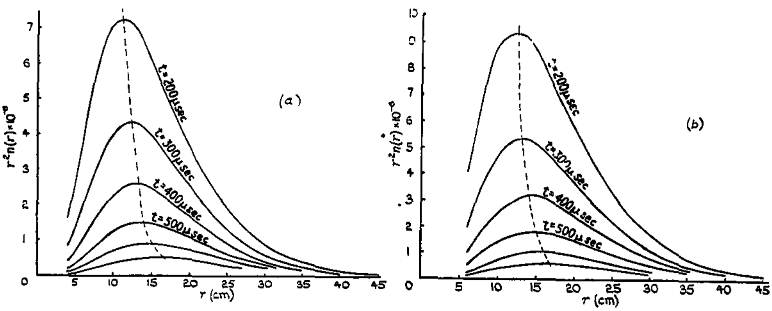

Figure 2. Distribution of thermal neutrons about the target: (*a*) in the horizontal plane; (*b*) vertically below the target.

Slight deviations from strict exponential decay were observed in neutron measurements at a given position. The logarithmic plots of count against time were convex downwards for distances less than 10 cm from the target and convex upwards for greater distances. This effect was, however, only just discernible and did not prevent the assignment of a lifetime to each measurement. The variation with distance is shown in table 1.

Table 1

(1)	(2)	(3)	(4)	(5)	(6)
10	174·0 ± 0·6	171·7 ± 0·7	178·1	171·5	164·6
15	194·6 ± 0·4	192·6 ± 0·7	—	202·5	187·4
20	204·0 ± 0·2	201·7 ± 0·4	203·5	232·3	212·9
25	208·0 ± 0·84	—	—	261·0	236·0
30	211·1 ± 0·7	206·2 ± 0·85	211·8	278·0	249·3
35	207·4 ± 1·5	—	—	287·7	258·2
40	206·9 ± 0·8	205·3 ± 1·4	210·2	289·5	258·5
45	204·2 ± 0·7	202·2 ± 1·9	—	—	—
50	204·7 ± 0·5	201·9 ± 0·5	207·1		
55	203·1 ± 1·5	202·3 ± 1·8			
60	200·5 ± 0·85	197·6 ± 0·4			
65	201·5 ± 1·4	—			
70	203·7 ± 0·5	197·0 ± 0·8			
75	201·1 ± 1				
80	199·0 ± 1·4				

(1) Distance in cm of detector from target; (2) average life in μsec measured with γ counter in horizontal plane through target; (3) average life measured with γ-ray counter in horizontal plane through target with dummy target in position below target; (4) average life measured with γ counter vertically below target; (5) approximate average life measured with neutron counter in horizontal plane; (6) approximate average life measured with neutron counter vertically below the target

The errors quoted are the probable errors derived from the statistics. The neutron measurements have a negligible statistical error

The decay of the γ-ray scintillations was strictly exponential at all points in the tank at which it was measured. This result, which is rather surprising in view of the appreciable change in the neutron distribution caused by diffusion, was carefully verified. It is derived on simple diffusion theory by Collie, Meads and Lockett (1956). Typical decay curves are shown in figure 3. The result of all measurements can therefore be expressed graphically as the variation of average lifetime with position in the tank, and also the variation of counting rate in one channel with position. The effect of the dummy target was to reduce the average life by a few microseconds as shown in table 1.

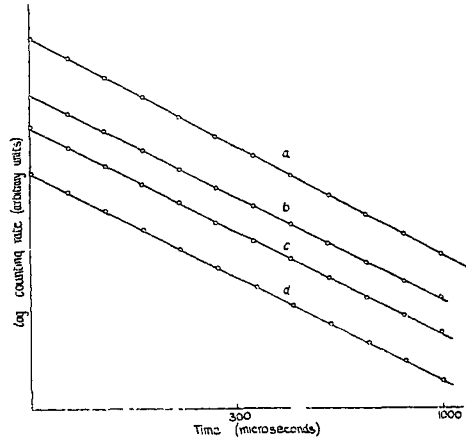

Figure 3. Exponential decay curves.

a γ-ray decay curve at 15 cm from the target in the presence of the dummy target $T=192{\cdot}6$ μsec;
b β-ray decay curve at 25 cm from the target $T=203{\cdot}3$ μsec;
c Integrated neutron curve $T=201{\cdot}9$ μsec,
d Integrated γ-ray curve $T=203{\cdot}3$ μsec

§ 9. The Angular Distribution of the Radiation

The neutrons emitted from the target are known to have a far from isotropic distribution even in the centre-of-gravity system; other factors causing a departure from symmetry are the presence of the target tube and the momentum of the incoming deuterons.

The initial lack of symmetry in the distribution of neutrons is of course greatly reduced by the random collisions of the fast neutrons in the slowing down process. The angular distribution was investigated mainly with a neutron counter. The results showed a peak in the backward direction near the target tube, but otherwise the neutron densities are well represented by a simple formula of the type $1 + a\cos^2\theta$. The existence of this backward peak is due to fast neutrons which have travelled up the target tube for some distance before entering the water. In this way a measurable concentration of neutrons is obtained near the target tube at distances from the target otherwise quite incompatible with the known stopping power of water for fast neutrons.

This appearance of neutrons near the free surface of the tank accounts for another puzzling feature of the experimental results, namely the rather large change in decay time (~ 5 μsec) introduced by the dummy target. It is possible to calculate from the measured neutron distribution how many neutrons leave the tank by diffusion up the target tube. It is quite small and corresponds to a target correction of $\frac{1}{3}$ μsec. The neutrons near the surface of the water tank will, however, be able to leave the tank by diffusion and so account for the larger dummy target effect. This explanation is confirmed by the observation that the dummy target correction is greater for large distances r from the target than for a small r, since the region near the point of entry of the beam into the tank subtends a proportionately larger solid angle as one proceeds outward from the target along a horizontal axis.

Since the neutron distribution determines the γ-ray distribution through a process of integration one would expect the latter to follow the same law but with a much smaller value of the coefficient of $\cos^2\theta$. For the purpose of calculation the γ-ray distribution was assumed to be of the form $1+a\cos^2\theta$ in which the coefficient a was determined from the already known counting rates at $\theta=0$ and $\theta=90°$ obtained from the measurements along the vertical and horizontal axes. An analysis of the considerable numerical data recorded by the delay sorter has been given by Meads (1955).

§ 10. Calculation of the Average Life

The probability of neutron capture was calculated from the experimental results in three ways:

(i) By integrating the γ-ray scintillations throughout the tank. Provided no neutrons and no γ-rays leave the tank the number of neutrons present is strictly proportional to the number of γ-rays captured in the tank, i.e. to the number of scintillations recorded, integrated over all positions with respect to the target. This integrated figure was obtained for each channel in each group, i.e. for 12 successive periods of 50 μsec from the experimental results in the following way.

The measurements beyond 35 cm from the target at a given time were accurately represented by the semi-empirical relationship $I_r=(I_0/r^2)e^{-kr}$ in which I_0 and k were determined experimentally.

This formula was used to extrapolate the results beyond the last point at which measurements were feasible. Any error introduced by this extrapolation was very small since it contributed only a few per cent of scintillations which all decayed with very nearly the correct average life. The effect of the target tube was allowed for by assuming that the difference between the ideal distribution with no target tube and the observed distribution would have been the same as the observed difference introduced by the dummy target. These are the only two corrections necessary to convert the observed measurements into the results of an ideal experiment in which a pulse of neutrons is released in an infinite volume of water. The integrated values for the various channels should show exponential decay. That this is so is shown in figure 3. The average life is found to be $203{\cdot}3\pm2{\cdot}6$ μsec. The estimated probable error is based on the following assumed errors: statistical error of the measurements 1 μsec uncertainty in the target correction (maximum correction is 6 μsec) 2 μsec an estimated uncertainty of 1–2 mm in the target–scintillator distance which would give an error of 1 μsec in the numerical integration.

(ii) The integration method can be applied to the neutron density as measured with the BF_3 chamber. This shows an excellent exponential decay (figure 3) with an average life of $201{\cdot}9\pm0{\cdot}13$ μsec. The error assigned is the statistical error of the measurements. There is also a systematic error due to the finite size of the counter and the difference between its capture cross section and that of the displaced water. Since the copper walled counters used have a higher absorption for neutrons than the displaced water one can expect (von Dardel and Waltner 1953) that the true value of the average life is greater than the observed 202 μsec.

(iii) The observed neutron distribution can be used to calculate the part played by diffusion in the observed exponential decay of the γ-ray scintillations in any part of the tank. The corrected value should then be the true average life due to capture alone and should be independent of the part in the tank for which it is calculated. A first order calculation (see Collie, Meads and Lockett 1956) gave the following corrections to the lifetime due to diffusion (table 2).

Table 2

(1)	(2)	(3)
12	+15·97	201·5
15	+ 2·87	197 8
20	− 8·27	198·5
25	− 11·31	202 3
30	-12 74	201 7
35	- 12·41	201 1
40	− 8·50	202 6
45	− 8 15	201 5
		Mean 200 7 μsec

(1) Distances from target (cm); (2) calculated correction (μsec), (3) corrected value of average life (μsec).

The diffusion correction behaves as was to be expected, being positive for small r when most of the neutrons are diffusing away from the detector and negative at large values of r when the neutrons are nearly all diffusing outwards towards the detector. A similar calculation, using the vertical data, was not carried out in full, since it became clear during the course of the above calculation, that tangential diffusion due to the asymmetry of the neutron distribution was not negligible. The calculation, which was very laborious, was unlikely to add much to the accuracy of the result as obtained by the first method. A value of the position in the tank of zero diffusion correction was, however, obtained from the vertical data. This was 18·3 cm, corresponding to a mean life of 204·0 μsec. It is likely, therefore, that a complete computation, taking the angular distribution of the neutrons into account, would yield a value not very different from 202 μsec.

We consider the first method (in which the total correction for angular distribution only amounts to a few microseconds) to be soundly based and the best way of treating the observational data.

The second and third methods give results which agree with the first method within their rather larger limits of error. This agreement while adding little to accuracy of the results is useful confirmation that the general picture of the behaviour of the expanding neutron cloud is correct.

§ 11. Comparison with Other Results

As has been seen the results of the measurements can be expressed in terms of a cross section for capture at a standard velocity (2200 m sec^{-1}) or as the average life in μsec of a neutron in water.

A summary of recent results expressed in barns is shown in table 3 which al indicates the method used to overcome the fundamental difficulties referre to in the introduction.

Table 3

Experimenters	Method	Results (barns) at 2200 m sec
Whitehouse and Graham (1947)	Static, boron ratio using small counters	Originally $0{\cdot}310 \pm 0{\cdot}025$. Wi revised boron value (760 barn $0{\cdot}332 \pm 0{\cdot}025$
Scott, Thomson and Wright (1954)	Dynamic, in small tank Escape correction measured by extrapolation	$0{\cdot}323 \pm 0{\cdot}008$
von Dardel and Waltner (1953)	Dynamic, in large tank using special BF_3 counters	$0{\cdot}321 \pm 0{\cdot}005$ (This result w discussed in the paper of v Dardel and Sjöstrand (195
Hamermesh, Ringo and Wexler (1953)	Static. Boron ratio using thermal neutrons and integrating tank. Boron comp. with standard	$0{\cdot}329 \pm 0{\cdot}004$
Harris *et al.* (1953)	Direct comparison with standard boron by pile oscillator method	$0{\cdot}332 \pm 0{\cdot}007$
von Dardel and Sjöstrand (1954)	Dynamic, in small tank. Escape correction obtained by extrapolation	$0{\cdot}333 \pm 0{\cdot}003$
Present Authors	Dynamic, in large tank, measuring the capture γ-rays	$0{\cdot}335 \pm 0{\cdot}004$

The present measurement is of the same order of accuracy as that of von Dard and Sjostrand (1954), who rely on the elimination of higher harmonic mode than the fundamental of the diffusion neutrons in a small tank of water. Th correction for 'buckling' in such a measurement involves extrapolation o measured lifetimes over a wide range of values.

This possible source of error is avoided by the present method of integratio of the capture γ-rays, which has the further advantage (previously discussed of avoiding the problem of the perturbing effect of BF_3 counters which arose in the static method and the work of von Dardel and Waltner (1953).

Acknowledgments

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