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Coherent Neutron-Proton Scattering by Liquid Mirror Reflection

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A measurement of the coherent neutron-proton scattering amplitude, utilizing total reflection of slow neutrons from a liquid hydrocarbon, was first reported in 1950. As the result ($-3.75\pm0.03\times10^{-13}$ cm) was significantly different from existing values, the measurements have been continued with a series of liquids in order to investigate systematic errors and to improve the accuracy. Final measurements have now been made utilizing a series of carefully purified hydrocarbon liquids, and a method in which no measurement of neutron wavelength is necessary. The ratio of the amplitude of hydrogen to that of carbon has been determined to 0.3 percent and the final value of the hydrogen amplitude, including the error (0.5 percent) in the carbon amplitude, is $(-3.78\pm0.02)\times10^{-13}$ cm (standard error).

I. INTRODUCTION

THE coherent neutron-proton scattering amplitude is one of the quantities which must be accurately known to fix the constants of low energy neutron-proton interactions. An accurate knowledge of the neutron-proton interactions allows a comparison with the corresponding proton-proton interactions, and this comparison in turn leads to important conclusions concerning charge dependence of nuclear forces. For some years the singlet $n-p$ force range was not determined with sufficient accuracy for comparison with the precisely measured singlet $p-p$ force range. An accurate comparison of $n-p$ and $p-p$ singlet ranges was of fundamental importance to nuclear theory, for inequality of these ranges would imply charge dependence of nuclear forces. In 1948 the $n-p$ singlet range, although of low accuracy, seemed to imply charge dependence.[1] Blatt and Jackson,[2] and Bethe,[3] in their reviews of the low energy scattering data, showed that, of the parameters that fix the singlet range, the one most in need of improved accuracy was the coherent $n-p$ scattering amplitude.

The coherent $n-p$ amplitude is difficult to measure, of course, because it constitutes only about 2 barns of a total bound atom cross section of 80 barns, the remainder representing spin-dependent incoherent scattering. The first measurements of the coherent $n-p$ amplitude were made by a study of interference effects in scattering of slow neutrons from ortho- and parahydrogen.[4] These measurements showed that the singlet state of the deuteron was virtual, but the value for the coherent amplitude combined with other scattering data implied an approximately zero singlet $n-p$ range. More accurate ortho-para measurements were made in 1947,[5] and these resulted in a coherent amplitude of $(-3.95\pm0.12)\times10^{-13}$ cm. The statistical accuracy of the measurements was reasonably high, but there was a strong possibility of systematic error resulting from the presence of ortho contamination in the parahydrogen. Because of the high cross section of the ortho-hydrogen relative to that of the para, a small amount of contamination would raise the observed amplitude appreciably. About the same time as the improved ortho-para measurements, the coherent amplitude was determined by Shull *et al.*[6] by the Bragg scattering of slow neutrons from crystals containing hydrogen. This diffraction method is quite straightforward, but suffers from the necessity of a correction for the reduction in observed coherent scattering by thermal vibrations of the crystal (Debye-Waller factor). The result of the crystal measurement was $(-3.96\pm0.20)\times10^{-13}$ cm in agreement with the ortho-para value. It was this value of 3.95 that was used by Blatt[1] in his 1948 calculation of the triplet and singlet force ranges. Blatt concluded that "this result (singlet range) is in direct contradiction to the commonly used hypothesis of the charge-independence of nuclear forces in the singlet state." It was clear that an accurate measurement of the coherent scattering was needed, especially in view of the possible systematic errors in the existing measurements.

II. FIRST EXPERIMENT

When the need for another measurement of the coherent scattering became clear, a repetition of the ortho-para experiment was considered at the Argonne National Laboratory. High intensities of velocity selected thermal neutrons were available, and it was certain that the actual cross-section measurements could be accurately made. It was clear, however, that the primary uncertainty in the experiment would not arise from statistical inaccuracy, but from the presence of ortho-hydrogen, and that there was no simple way of determining the ortho-contamination in the parahydrogen. About the same time at Argonne, Hughes

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[1] J. M. Blatt, Phys. Rev. **74**, 92 (1948).

[2] J. M. Blatt and J. D. Jackson, Phys. Rev. **76**, 18 (1949).

[3] H. A. Bethe, Phys. Rev. **76**, 38 (1949).

[4] Halpern, Estermann, Simpson, and Stern, Phys. Rev. **52**, 142 (1937); Brickwedde, Dunning, Hoge, and Manley, Phys. Rev. **54**, 266 (1938); L. W. Alvarez and K. S. Pitzer, Phys. Rev. **58**, 1003 (1940).

[5] Sutton, Hall, Anderson, Bridge, DeWire, Lavatelli, Long, Snyder, and Williams, Phys. Rev. **72**, 1147 (1947).

[6] Shull, Wollan, Morton, and Davidson, Phys. Rev. **73**, 842 (1948).

and Burgy[7] had shown in a series of mirror experiments that the critical angle for total reflection is related to the coherent scattering in an unambiguous way. For instance, the critical angle for an unmagnetized iron mirror is unaffected by magnetic scattering even though the iron is composed by domains (about 10^{-3} cm in extent) that are completely magnetized. The critical angle is determined by the average coherent scattering amplitude, and the magnetic amplitude, when averaged over the surface of the mirror, becomes zero, leaving only the nuclear amplitude. Because of this averaging property of a mirror, the critical angle could be measured for a mixture of substances (liquid, solid, or gas) and the coherent amplitude obtained would be an accurate average of the amplitudes of the constituents, regardless of their molecular form or crystalline state. The coherent amplitude obtained with a mirror, furthermore, is not affected by temperature vibration of the material, hence there is no Debye-Waller correction to be made, as in the diffraction method. The essential reason for the disappearance of the Debye-Waller correction is that the index of refraction and the critical angle are determined by the scattering in the forward direction.[8]

It was suggested by Hamermesh and Wattenberg[9] that the advantages of the mirror technique would be of great value for the hydrogen scattering, in which the coherent scattering is so much larger than the coherent. Reflection of neutrons from a hydrogen mirror would give the coherent amplitude directly with no interference from the incoherent scattering. However, the required accuracy in the hydrogen amplitude (about 1 percent) could not be obtained by a direct measurement of reflection from hydrogen. Further difficulty would be encountered because the negative scattering amplitude would necessitate internal reflection and a consequent serious loss in intensity. Some months later, however, a method was devised that would overcome the difficulties of reflection from hydrogen itself. The method consists of over-balancing the hydrogen amplitude slightly by adding carbon which has a positive, accurately known amplitude. External reflection is then used for the mixture and, as only the unbalanced amplitude is measured, the required accuracy is greatly reduced.

The results of the first measurements with the balancing method were reported in a Letter to the Editor,[10] and the details will not be repeated here. Neutrons were reflected from a liquid, triethylbenzene, and the limiting wavelength reflected was measured by transmission through gold and by reflection at a beryllium error. The amplitude of the liquid is then given by

$$\theta=\lambda(N\Delta a/\pi)^{\frac{1}{2}}, \tag{1}$$

where θ is the incident angle, λ the limiting wavelength reflected, and N the number of carbon atoms per cm^3. The amplitude Δa represents the net or unbalanced amplitude per carbon atom and its value gives the hydrogen amplitude in terms of that of carbon. The experiment was not difficult to perform to the planned accuracy of one percent and was free from any uncertainties in interpretation. The result, however, was $-(3.75\pm0.03)\times10^{-13}$ cm which value disagreed with the ortho-para and diffraction results by more than the errors involved.

III. SECOND EXPERIMENT

Although no source of systematic error was known, it was decided to extend the measurements to investigate the possibility of such errors, and to increase the accuracy. The possibility of systematic errors could be investigated by repeating the measurement for various liquids and by using different neutron wavelengths. An increase in the accuracy beyond the one percent already obtained necessitated a change in method to eliminate effects which could be circumvented in the first experiment but which would prevent any significant increase in accuracy. The natural way to increase the accuracy would be to use a liquid in which the hydrogen amplitude was more nearly balanced by the carbon. For such a liquid the limiting wavelength reflected would be longer if the incident angle remained the same as shown by Eq. (1). With thermal neutrons this meant very low intensities since even in the first experiment the wavelengths involved were well below the maximum of the thermal spectrum. The intensities were so low that transmission and second reflection experiments were considered impractical. It was therefore decided to use the intensity itself as a measure of the critical wavelength. On the low velocity side of the maximum of the distribution of thermal neutrons, the reflected intensity depends on the fourth power of the critical wavelength if the distribution is maxwellian. From Eq. 1 it can be seen that at a given angle the intensity depends then on the square of the residual amplitude, and hence is a fairly sensitive measure of it. A plot of the square root of the intensity at a given angle for a number of hydrocarbons *vs* the hydrogen to carbon content ratio should thus cross the zero ordinate at the ratio where the two amplitudes balance. In this method it is assumed that the reflectivity of the liquid surfaces is nearly the same for all liquids for neutrons of greater than critical wavelengths. Measurements on liquids of known amplitude bear out this assumption.

At long wavelengths, however, the neutron distribution is no longer maxwellian, and the method just described becomes increasingly difficult to interpret. As the neutron wavelength increases above about 5A the intensity drops far more rapidly with wavelength than

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[7] D. J. Hughes and M. T. Burgy, Phys. Rev. **81**, 498 (1951).

[8] It was suggested recently by D. Kleinman and G. Snow, Phys. Rev. **82**, 952 (1951), that there might be a correction as large as 1 percent in the liquid mirror experiment, arising from virtual inelastic scattering. Further calculation by Kleinman and Snow shows that the correction is probably negligible (private communication).

[9] M. Hamermesh, Phys. Rev. **77**, 140 (1950).

[10] Hughes, Brugy, and Ringo, Phys. Rev. **77**, 291 (1950).

expected for a maxwellian distribution. At 6.6A units the intensity increases discontinuously as the wavelength becomes longer than the largest lattice spacing in the graphite of the pile, and at still longer wavelengths it continues to decrease faster than the maxwellian. Another effect that becomes increasingly important with wavelength is the finite reflectivity for neutrons of wavelength less than the limiting wavelength. This "spill-over" effect becomes so large with wavelength, that for limiting wavelengths in the 10–15A range, approximately as many neutrons with wavelengths shorter than critical are reflected as those of wavelength longer than critical. Because of these effects it was clear that an improvement in the experiment must involve a method that would avoid any dependence on an accurate knowledge of the wavelength distribution of the incident neutrons.

The method of measurement finally adopted in these experiments avoided the dependence on knowledge of the distribution in the following way. Even though the spectrum is not maxwellian, it decreases sharply in intensity with increasing wavelength, so the reflected intensity (which is proportional to the integral from ∞ wavelengths up to the critical wavelength), decreases even faster, and is a very sensitive function of wavelength. Thus a given intensity of reflection means a given critical wavelength. The experiment consists of measurements of those values of the incident angle that give constant intensity (i.e., a constant critical wavelength) in the reflected beam for several liquids of different hydrogen/carbon content ratios. As can be seen from Eq. 1, for a given wavelength the square of the incident angle is proportional to the net amplitude, which in turn is a linear function of the H/C ratio. A plot of θ^2 *vs* H/C content should then be a straight line reaching $\theta^2=0$ at the point when the H/C ratio is the negative inverse of the ratio of the coherent scattering amplitudes of H and C.

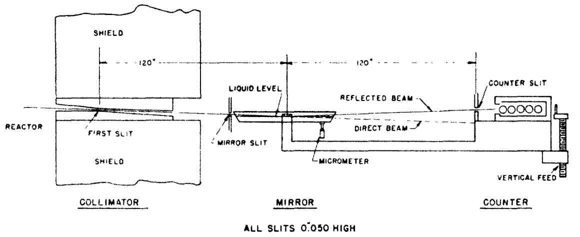

FIG. 1. Apparatus for neutron reflection from liquids.

This method is very simple to use, involves no measurement of the wavelength, works on any reasonable spectrum shape, and is even valid if the reflection is dominated by spill-over. This last property can be seen from the equation for reflectivity beyond the critical angle:

$$\text{Reflectivity} = \tfrac{1}{16}(\theta_c/\theta)^4 \quad \text{for} \quad \theta_c/\theta \gg 1.$$

Here θ_c is the largest angle at which a wavelength λ is totally reflected as given by Eq. (1). This result shows that the spill-over intensity decreases rapidly as the critical wavelength increases, so that again the intensity is a sensitive measure of the critical wavelength.

IV. APPARATUS

The experimental arrangement at the thermal column of the Argonne heavy water moderated reactor is shown roughly in Fig. 1. It is quite similar to that of the first experiment with a few exceptions. The second mirror is absent and a considerably longer mirror tray was used, giving a liquid surface 48 in. long. The ends of this tray were tilted 60° from the vertical so that the meniscus would not extend above the surface of the liquid and interfere with the beams. The counter slit and counter were mounted on a steel frame, which was pivoted at a point near the center of the mirror surface so that the beams always traversed the counters at nearly the same place. The mirror tray was made of stainles steel with windows of 0.005 in. stainless steel seam welded in the ends of the tray. The tray was provided with a gasket and a glass cover to minimize evaporation of the mirror liquid. No special precautions were taken to damp out surface waves in the mirror. It was found that the reflected beam was not appreciably wider than the direct beam even when there were distinctly visible waves on the mirror surface with several large fans and pumps running within 50 ft of the mirror.

The mirror and counter slits consisted of machined blocks of cadmium held on an aluminum frame which could be readily adjusted in height and level. The collimator was made of graphite with a cadmium-plated steel lining. It was designed so that fast neutrons would have to pass through at least 2 in. of steel and 1 ft of graphite to go directly from the first slit to the counter. The collimator and the slits were 4 in. wide in the direction perpendicular to the plane of Fig. 1. The counter consisted of a bank of $5B^{10}F_3$ counters which were nearly "black" to neutrons in the energy region of interest in these experiments. A fission counter used as a monitor was placed in the shield at the side of the collimator.

The liquids used in the measurement were triethylbenzene, $C_6H_3(C_2H_5)_3$, cyclohexene, C_6H_{10}, and a mixture of benzene and cyclohexane, C_6H_{12}, designed to give a hydrogen to carbon atomic ratio of 1.700. In the first set of experiments using the present method, it was found that the principal uncertainty in the measurement lay in the ratio of the hydrogen to carbon content of the liquids. The experiments were therefore repeated, taking considerable pains with this point. The best commercial grades of the hydrocarbons were obtained and these were carefully repurified by fractional distillations. As a check, however, quantitative analyses of this material were run by Ralph Bane of the Argonne Chemistry Division. To make the measurements conveniently about 400 cc of each liquid were needed.

V. MEASUREMENTS

A. Angle

The direct beam was first located with the mirror tray removed, then the mirror tray was put into place, the liquid poured in, and the reflected beam located and its intensity measured. An example of such a measurement is shown in Fig. 2. The incident angle is, of course, given by half the difference in the positions of direct and reflected beam, divided by the distance from the counter slit to the point on the mirror surface where the center of the beam is incident.

The most difficult part of the angle measurement is the determination of the incident point on the mirror. It was done by making a series of measurements of the reflected beam intensity as a function of mirror height for a given setting of the mirror slit, i.e., a given angle of incidence. When the incident beam over-shoots the far end of the mirror the intensity drops, similarly when the beam undershoots the near end, the intensity drops. A plot of the reflected intensities (always measured at the peak of the reflected beam) as a function of mirror position is shown in Fig. 3. It was assumed that the center of the beam was incident on the center of the mirror surface when the mirror height was halfway between the two heights where the intensity was half that of the plateau. This was a rather tedious measurement, but fortunately it was only necessary to do it once as all other angles could be determined relative to the measured one by noting the change in the position of the mirror slit, or the direct beam at the counter slit, in going to the new angle. For speed and accuracy the reference angle measurement of Fig. 3 was made using carbon tetrachloride in the mirror as it is a good reflector. In actual practice both the mirror slit position and the direct beam measurement were used to give the change in angle and agreed within the accuracy of each—about 0.1 min. At the end of the measurements the position of the direct beam was always checked by removing the mirror and it was always found in the original position, indicating that the apparatus was sufficiently stable.

B. Intensity

For the intensity measurements on the various liquids it was necessary only that the mirror be in a position on the plateau of Fig. 3 and this was not a critical adjustment. It could in fact be determined simply from the position where the mirror cut off the direct beam. The measurements were, of course, made on the peaks of the reflected beams, and for comparison were reduced to a given number of counts from the monitor counter.

In general, the backgrounds were not obtained by putting cadmium in the beam, but rather by interpolating the intensities on either side of the beam. This procedure was necessary for there was a small background of scattered thermal neutrons, whose intensity decreased fairly rapidly away from the direct beam but was still quite appreciable at the smaller angles of reflection. This small thermal neutron background could not be excluded in spite of careful placing of cadmium slits to define the beam.

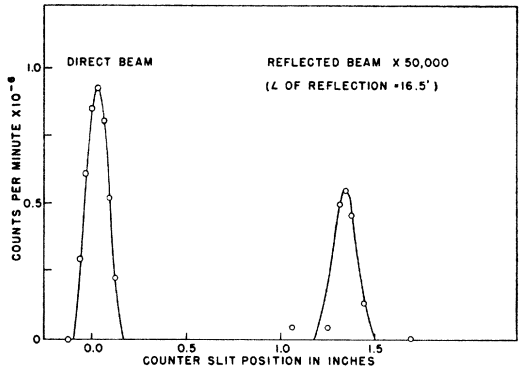

FIG. 2. Direct beam and beam reflected from triethylbenzene mirror. The zero on the position scale is arbitrary.

In the measurement on the liquids mixed to give a 1.700 H to C content ratio, the possibility of differential evaporation was checked by observing the intensity reflected from the mirror over a period of hours, and no change was observed. For brevity the description of the experiment has omitted the discussion of a number of small corrections. We have, however, made a correction for the density of the mirror liquid, and we have considered and found negligible, corrections for the change in wavelength resolution of the mirror with angle, and the change in position of the neutron path inside the counter with incident angle.

VI. RESULTS

The results of the measurements on the three liquids are given in Table I and the points obtained from these results are shown in Fig. 4. As can be seen from Table I, the incident angles were not set to give precisely equal intensities for the different liquids, and the values of θ^2 in Fig. 4 were adjusted to equal intensities (as well as to equal densities). Figure 4 gives a value of the ratio of the carbon amplitude to the hydrogen amplitude

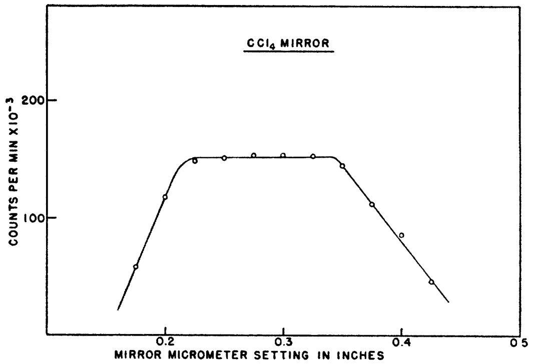

FIG. 3. Reflected intensity as a function of the height of the mirror surface. Thus curve is used to center the beam on the mirror.

TABLE I. Intensity of neutrons reflected from various liquids as a function of incident angle. The limiting wavelength for total reflection is included, although it is not necessary in the analysis (Fig. 4).

Liquid	Angle minutes	Intensity in counts per min*	λ(calculated) angstroms
Triethylbenzene	16.5	130	13.5
	14.6	244	12.3
	12.7	465	10.7
Cyclohexene	9.50	121	14.2
	8.33	185	12.5
	7.02	451	10.5
1.700 mixture	7.02	129	13.6

* Here and in the figure the intensity is normalized to a monitor rate of 17,000 cpm.

(a_C/a_H) of -1.748 ± 0.005. The error is estimated largely from the variation of the ratio deduced from measurements at different intensities, and from the differences between the formulas and analyses for the H/C ratio. Including the results of the first experiment, with appropriate weighting, we obtain finally

$$a_C/a_H = -1.753\pm0.005.$$

To obtain the value of a_H we must know the value of a_C. The transmission measurements on carbon give the total cross section to quite high accuracy $\sigma_C = 4.70 \pm 0.05$[11] from 1 to 500 ev. If there is no incoherent scattering, $a_C = (13/12)(\sigma/4\pi)^{\frac{1}{2}}$. Since the capture cross section is negligible and the spin of C^{12} is zero, the only uncertainty is obtaining a_C from σ_C is the cross section of C^{13}. Recent studies at Brookhaven[12] of thermal incoherent scattering show that the isotopic plus spin-incoherent scattering is less than 0.1 barn. This result in turn shows that the scattering amplitude of C^{13} times its isotopic abundance is not more than 1 percent that of C^{12}. In addition the total absence of any indication of a resonance in the neutron cross section of carbon below 5000 ev makes it quite plausible that C^{13} has a positive scattering amplitude and one within a factor of two that of C^{12}. It then follows that $a_C = 6.63(\pm0.03) \times 10^{-13}$ cm and $a_H = -3.78(\pm0.02)\times10^{-13}$ cm.

As can be seen from the errors quoted, we now believe this measurement of the ratio of the coherent scattering amplitude of carbon and hydrogen to be more accurate than the current values for the amplitude of carbon. It seems, however, fairly straightforward to improve this last to about the same error as that for the ratio. The fact that the different liquids and different critical wavelengths all give values in quite good agreement, as shown by Fig. 4, encourages the belief that there are no important systematic errors in this measurement of the ratio.

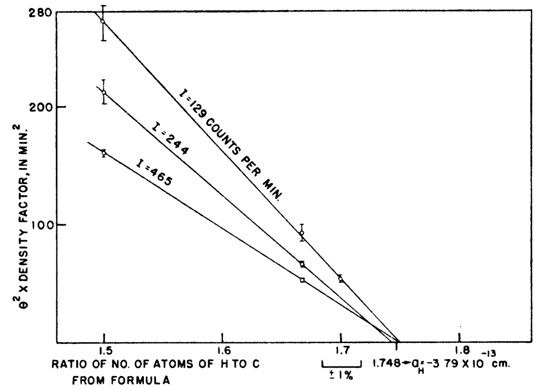

FIG. 4. Graph for determination of balance point of H to C coherent neutron scattering amplitudes.

The value for a_H quoted, $-3.78(\pm0.02)\times10^{-13}$ cm, together with Melkonian's[13] value for the free proton cross section 20.36(±0.10) barns, gives the triplet and singlet amplitudes:

$$a_T = 5.377(\pm0.023)\times10^{-13} \text{ cm}$$

and

$$a_S = -23.69(\pm0.06)\times10^{-13} \text{ cm}.$$

The triplet amplitude plus the Mobley and Laubenstein[14] value for the binding energy of the deuteron, 2.226(±0.003) Mev, gives the effective range of the triplet well:[15–17]

$$\rho_t = 1.704(\pm0.030)\times10^{-13} \text{ cm}.$$

These values do not differ significantly from those used[10] in several papers on the comparison of neutron-proton and proton-proton forces,[16–18] the mass of the π-meson,[19] the binding energy of the triton[20] and the shape of the neutron-proton interaction potential.[21,17] It can be seen from reference 17 that within the limits of error, which now appear to come mainly from the 1- to 5-Mev neutron-proton cross-section measurements, the derived $n-p$ and $p-p$ singlet ranges are consistent with charge independence of nuclear forces.

It is a pleasure to acknowledge the considerable help we received on theory from Dr. Morton Hamermesh and Dr. Hartland Snyder, and on chemistry from Dr. Solomon Wexler.

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