

and 1379 kev, it is difficult to escape the impression of the presence of a typical fine structure.

In a forthcoming publication from this institute it is planned to give a more detailed discussion of the characteristics of the spectrum.

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Interference Phenomena of Slow Neutrons

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Various experiments involving interference of slow neutrons have been performed in order to determine the phase of the scattered neutron wave with respect to the primary neutron wave. Theoretically this phase change is very close to either 0° or 180° . The experiments show that with few exceptions the latter is the case. The evidence is based on the following types of measurements: (a) measurement of the intensities of Bragg reflection of various orders of many crystals, and comparison with the theoretical values of the form factor; (b) total scattering cross section of gas molecules for wave-lengths long compared with the molecular dimensions; and (c) determination of the limiting angles for total reflection of neutrons on various mirrors. The elements Ba, Be, C, Ca, Cu, F, Fe, Mg, N, Ni, O, Pb, S, and Zn were found to scatter neutrons with 180° phase difference; Li and probably Mn scatter with zero phase difference. The five elements I, Br, Cl, K, and Na behave alike and the phase with which they scatter is tentatively identified as 180° . Coherent scattering cross sections have been determined for several elements.

INTRODUCTION

THE scattering processes of slow neutrons are greatly complicated by interference phenomena due to the fact that the de Broglie wave-length is comparable with interatomic distances. The general pattern of interference phenomena of slow neutrons is similar to that of x-rays, since both the wave-length and the scattering cross section of x-rays are comparable to those of neutrons. On the other hand there are considerable differences due to several factors. Among them is the fact that the scattering of x-rays varies regularly with atomic number while that of neutrons is a rather erratic property. Furthermore, in the case of neutrons the phase difference between scattered and incident wave may be either 0° or 180° as will be discussed in Section 1. For x-rays instead, it is always 180°

because x-ray energies are larger than most electronic resonance energies. Also, the absorption properties of neutrons differ markedly from those of x-rays.

The main purpose of this work was the investigation of various interference phenomena in order to determine the phase change of the scattered neutron wave for a large number of elements. Section 1 contains a summary of the theoretical background of this work. Section 2 describes the measurements of the intensities of Bragg reflections of various orders and their interpretation. Section 3 is a discussion of some experiments on filtered neutrons. In Section 4, experiments on scattering of neutrons by gas molecules are presented. Section 5 describes measurements of the limiting angle for total reflection of neutrons. The general conclusions are discussed in Section 6.

1. Theoretical Considerations

When a slow neutron is scattered by a nucleus, its de Broglie wave at some distance from the scattering nucleus may be written as the sum of a term $\exp(ikx)$ representing the primary wave and a term $-a[\exp(ikr)]/r$ representing the scattered wave. This is true for the case of slow neutrons because with very good approximation the scattering is spherically symmetrical. The coefficient of the latter term has been written with the minus sign for reasons of convenience that will be apparent later. If the constant a is positive, there is a phase change of 180° between scattered and incident wave, and if a is negative, the phase change is 0° . One proves in an elementary way that the scattering cross section is related to the constant a by the equation

$$\sigma = 4\pi |a|^2 \quad (1)$$

and also that the constant a is with very good approximation for slow neutrons a real number. Its imaginary part is very small and can usually be neglected except in case of extremely high absorption. The quantity a which has the dimensions of a length and the order of magnitude of 10^{-12} cm shall be referred to as the "scattering length." The main purpose of this paper is the experimental determination of the scattering length and in particular of its sign.

For elements consisting of one isotope only and without nuclear spin, the magnitude of the scattering length can be immediately obtained from (1), so that only its sign needs to be determined. Even in this simple case a small correction must be applied depending on whether the atom is free or bound. Because of the change in the reduced mass, the cross section of a free atom differs from that of the same atom bound in a crystal, by a factor $[(A+1)/A]^2$, where A is the atomic weight.¹

Since the scattering length is proportional to the square root of the cross section, the correction factor will be $(A+1)/A$. It follows that

$$a = [(A+1)/A] a_f \quad (2)$$

where a indicates the scattering length of the atom bound in a crystal and a_f the scattering

length for the free atom. For elements that are mixtures of several isotopes with probabilities of occurrence p_1, p_2, \dots, p_n , and scattering lengths a_1, a_2, \dots, a_n the scattering length is the average

$$a = p_1 a_1 + p_2 a_2 + \dots + p_n a_n. \quad (3)$$

This is the magnitude that determines the interference properties of the element. The scattering cross section is no longer given by (1) but by

$$\sigma = 4\pi \{p_1 a_1^2 + p_2 a_2^2 + \dots + p_n a_n^2\}. \quad (4)$$

Actually in this case the relationship between a and σ can be expressed by the inequality

$$|a| \leq (\sigma/4\pi)^{1/2}. \quad (5)$$

The knowledge of the scattering cross section is insufficient, therefore, to determine even the magnitude of the scattering length.

A similar situation obtains if the nucleus has a spin I different from zero. Here the spin vector of the scattering nucleus is oriented either parallel (total spin $I + \frac{1}{2}$), or antiparallel (total spin $I - \frac{1}{2}$) to the spin vector of the neutron. Consequently the scattering length may take either of two values, $a_{I+\frac{1}{2}}$ or $a_{I-\frac{1}{2}}$. The effective scattering length is the average of these two scattering lengths, each being weighted by the probability of occurrence of the corresponding spin orientation, namely

$$a = \frac{I}{2I+1} a_{I-\frac{1}{2}} + \frac{I+1}{2I+1} a_{I+\frac{1}{2}}. \quad (6)$$

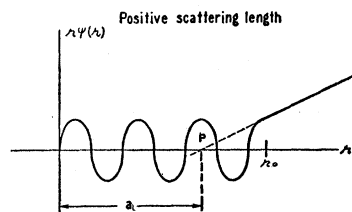


FIG. 1A. Positive scattering length.

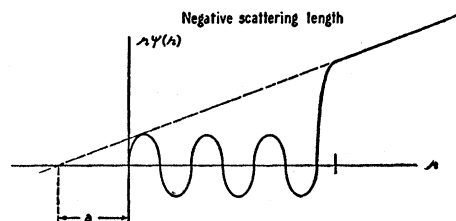


FIG. 1B. Negative scattering length.

¹For example see H. A. Bethe, Rev. Mod. Phys. 9, 71 (1937).

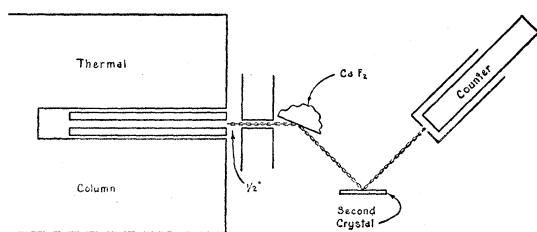


FIG. 2. Apparatus for measuring intensity of Bragg orders.

The scattering length will obey (5), the equal sign corresponding to the case that $a_{l+\frac{1}{2}}$ is equal to $a_{l-\frac{1}{2}}$ both in magnitude and sign.

A simple geometrical interpretation of the scattering length can be obtained if the interaction between the neutron and the nucleus is represented by a potential well. Let the eigenfunction of the neutron for an s state of energy zero be $\psi(r)$. In Fig. 1, $r\psi(r)$ is plotted *versus* r . The curve becomes a straight line for r greater than the radius r_0 of the potential well. This straight line can be extended to intersect the r axis at the point P . One can prove that the scattering length a is the abscissa of the point P . Figure 1A corresponds to a case in which a is positive, and Fig. 1B to a case in which a is negative. By inspecting the two figures it is apparent that the probability is much higher for a positive than for a negative, especially for heavy elements. This remark on the relative probability for a to be positive or negative is due to E. Teller and V. Weisskopf.

A simple formula can be obtained in the case that the scattering is due to the effect of a single Breit-Wigner resonance level. It is, then

$$a = -\lambda_R \Gamma_n / [W - R + i\Gamma] \quad (7)$$

where W is the energy of the neutron, R is the resonance energy, Γ_n and Γ are the neutron and total half width at half maximum, and λ_R is the de Broglie wave-length at energy R , divided by 2π . Since in most cases Γ is small compared with $(W - R)$, it follows that a will be positive or negative depending on whether the energy of the neutron lies below or above the resonance energy. This picture is a great oversimplification since there are no cases in which the Breit-Wigner scattering of a single level is the dominant phenomenon, and complicated situations arise

due to interference of Breit-Wigner and potential scattering.

2. Intensities of Various Orders of Bragg Reflection

One can compare the changes in phase in the neutron scattering of different elements by measuring the intensity of the Bragg scattering for various orders and various crystalline planes of crystals containing at least two elements.

The simplest case is that of a crystal in which the planes are equidistant and consist alternately of two kinds of atoms, as for example, the 1, 1, 1 planes of NaCl. These planes are equidistant and consist alternately of sodium and of chlorine. In the first order Bragg reflection, the optical path for reflection from sodium planes differs from the path for reflection from chlorine planes by $\lambda/2$. Consequently if sodium and chlorine nuclei cause the same change in phase of the scattered neutron wave, their contributions will subtract and the order will have low intensity. If they scatter with opposite phase change their contributions will add and the order will have high intensity.

The situation for the second order is reversed. Here the difference in optical path for reflection from the two kinds of planes is λ . Consequently if sodium and chlorine scatter with the same change in phase the order will have high intensity, and if with opposite change in phase, low intensity. Similarly it follows that if the two kinds of nuclei scatter with the same change in phase the odd orders will be weak and the even orders will be strong. Conversely, if they scatter with opposite change in phase, the odd orders will be strong and the even orders weak.

Analogously to x-ray scattering, there is superimposed upon this effect a continuous decrease in intensity from order to order due to geometrical factors, to thermal agitation, and to imperfections of the crystal.

For more complicated cases the intensity of the various orders is determined as for x-rays by the form factor

$$F = \left| \sum_j a_j \exp(2\pi i n d_j / d) \right| \quad (8)$$

where a_j is the scattering length, d is the spacing of the lattice planes, n is the order of the Bragg

reflection, and δ_j is the perpendicular distance from the j th atom to the plane of reflection. Naturally the form factor is strongly dependent on the relative signs of the scattering lengths of the various atoms of the crystal. An analysis of the intensities for various orders and various planes of the crystal will often allow the determination of the relative signs of the scattering lengths of the elements involved. In several cases it is possible to determine also the ratios of the values of the scattering lengths.

A comparison of intensities for Bragg reflections of various orders of many crystals have been made using monochromatic neutrons. The experimental arrangement is schematically represented in Fig. 2. A beam of non-monochromatic thermal neutrons emerges from a $4'' \times 4''$ hole containing a long collimator $\frac{1}{2}''$ wide by $1\frac{1}{4}''$ high in the thermal column of the Argonne heavy water pile. This beam falls on the 1, 0, 0 plane of a large CaF_2 crystal at a glancing angle of about 16° . Neutrons of energies satisfying the Bragg condition ($\lambda \sim 1.50$ angstroms for the first order) are reflected to form a beam that is used for investigating various crystal specimens. The beam contains a main component of neutrons of wave-length 1.50\AA plus a small fraction of neutrons of energy four times as large due to the second order reflection on the CaF_2 crystal. The second order component is fairly unimportant because there is only a small number of neutrons of this relatively high energy in the Maxwell distribution. We can, therefore, consider the beam reflected on the calcite to be approximately monochromatic. This monochromatic beam of neutrons falls on some plane of the second crystal, which is mounted on a rotating table. The neutrons undergo a second Bragg reflection, and are finally detected by an enriched BF_3 proportional counter. The counter is supported by an arm which rotates about the axis of the crystal table. Additional Cd slits not shown are used for more precise collimation of the neutron beam.

A typical measurement was carried out as follows. Both counter and second crystal were set approximately at angles corresponding to a reflection of a given order. By rocking each separately the setting of maximum intensity was found. Also the inclination of the crystal was adjusted for maximum intensity. From the in-

tensity measured in these conditions was subtracted a background, measured by turning the crystal a small angle off the Bragg position. In the majority of cases the background was a small fraction of the total count.

The results of these measurements are summarized in Table I. Columns 1 and 2 indicate the crystal and the plane. Column 3 is the form factor. Here the chemical symbol of the element has been used to represent its scattering length. The fourth column gives the absolute value of the form factor, using the values of the scattering length given in Table VII. The fifth column gives the measured intensities of the various orders in counts per minute. In the sixth column is the ratio of column 5 to column 4. If the values chosen for the scattering lengths are approximately correct, one expects that the values in column 6, corresponding to any given plane, should show a regular decrease in intensity with increasing order, since dividing the intensity by the form factor should correct it for the irregular change of intensity from order to order. Therefore only the regular decrease should remain.

There may be some doubt whether it is more appropriate to divide the intensity by the form factor or by its square. Theoretically one would expect that, for ideally perfect crystals, the form factor should be used, and for ideally imperfect crystals, its square. Actually, we have found that one obtains a much better fit by using the form factor.

For the simplest crystals containing two elements one can see immediately by inspecting the intensity data of column 5 whether the scattering lengths of the two elements have equal or opposite sign. For instance CaF_2 (100), NaCl (111), and PbS (111) show clearly superimposed on the general decrease of intensity with increasing order, an alternation of intensity with strong even orders and weak odd orders. Since, in these crystals, the planes consist alternately of the two kinds of atoms, we conclude that the scattering lengths of each pair have the same sign. The opposite is the case for LiF (111) where the even orders have intensity so low that we could not measure them, and the odd orders have a normal intensity. Therefore Li and F have scattering lengths of opposite sign. In some of the more complicated cases the analysis is also

TABLE I. Intensity of Bragg orders.

Crystal	Plane	Order	Form factor		Intensity	Ratio
CaF ₂	(100)	1	Ca-2 F	.41	16300	39800
		2	Ca+2 F	1.99	20300	10200
		3	Ca-2 F	.41	1287	3140
NaCl	(111)	1	Na-Cl	.57	2376	3990
		2	Na+Cl	1.69	2750	1630
PbS	(111)	1	Pb-S	.20	7280	36400
		2	Pb+S	.76	10700	14100
		3	Pb-S	.20	808	4040
		4	Pb+S	.76	750	986
PbS	(100)	1	Pb+S	.76	19650	25800
		2	Pb+S	.76	11420	15000
		3	Pb+S	.76	2249	2960
FeS ₂	(100)	1	Fe+ .62 S	.99	6893	6960
		2	Fe-1.6 S	.37	1354	3660
		3	Fe-1.6 S	.37	726	1960
MnS ₂	(111)	1	Mn-1.06 S	.74	7930	10700
		2	Mn+ .06 S	.42	2560	6100
		3	Mn+ .06 S	.42	670	1600
Fe ₃ O ₄ (magnetite)	(111)	1	2.34 Fe+ 1.20 O	2.65	21300	8040
		2	16 Fe-31.9 O	6.34	19970	3150
		3	13.7 Fe- 3.55 O	9.06	12570	1390
		4	8 Fe+31.50 O	25.78	16700	647
		5	13.7 Fe+ 5.94 O	14.85	5120	344
CaCO ₃ (calcite)	(111)	1	2 C+6 O-2 Ca	3.42	9500	2780
		2	2 C+6 O+2 Ca	6.58	8520	1290
		3	2 C+6 O-2 Ca	3.42	1840	538
CaCO ₃ cleavage	(211)	1	2 Ca+2 C+1.76 O	3.99	10900	2730
		2	2 Ca+2 C-2.0 O	1.70	3360	1980
		3	2 Ca+2 C+2.7 O	4.57	2400	528

quite straight forward as can be seen by comparison of the observed intensities with the formulae for the form factors in column 3. In a few cases the interpretation is not unique.

The following conclusions as to the signs of the scattering lengths can be drawn. The components of the following pairs have scattering lengths of the same sign: Na, Cl; Pb, S; Ca, F; Fe, S; Fe, O; K, Br; Mg, O; K, Cl; K, I. The components of the pair, Li, F have scattering lengths of opposite sign. The measurements of MnS₂ do not allow a unique interpretation although the probable conclusion is that Mn and S also have scattering lengths of opposite sign.

Using the evidence to be presented in Sections 4 and 5 that carbon and oxygen have scattering lengths with the same sign, one can conclude from the measurements on calcite that Ca and O also have scattering lengths of the same sign.

From the measurements on FeS₂ and Fe₃O₄,

one can conclude that S and O have the same sign; and from data on BaSO₄, one is led to assign also the same sign to Ba. The measurements on NaNO₃ indicate that N and O have the same sign. (See confirmatory evidence in Section 4.) The measurements of the (111) plane of NaNO₃ indicate although not quite conclusively that Na has the same sign as the group NO₃, and therefore as N and O.

From these data it follows that C, O, Fe, Mg, Ba, Ca, S, F, Pb, and N all have the same sign. Na, K, Cl, Br, I all have like sign. The partial evidence just mentioned, according to which Na has the same sign as O, indicates that all this latter group should be included with the former. That this conclusion is correct is strongly supported by evidence to be presented in Section 6. Li and perhaps Mn have scattering lengths of the other sign. They represent together with hydrogen (see Section 6) the only exceptions

TABLE I.—Continued.

Crystal	Plane	Order	Form factor	Intensity	Ratio
BaSO ₄	(100)	1	4 Ba+4 S+ .42 O 4.54	3606	794
		2	4 Ba+4 S+14.6 O 13.19	4837	366
		3	4 Ba+4 S+ 3.1 O 6.17	756	123
BaSO ₄	(011)	1	-3.4 Ba+3.8 S+4.7 O 1.24	1018	821
		2	1.75 Ba+3.1 S-9.6 O 3.61	1632	452
		3	.42 Ba+2.0 S-7.9 O 3.93	651	166
		4	-2.46 Ba+ .75 S-2.2 O 3.07	36	12
BaSO ₄	(101)	1	.87 Ba-1.25 S- 1.47 O .56	737	1320
		2	1.13 Ba-2.02 S- 4.56 O 2.46	1946	791
		3	3.77 Ba+ .98 S+ 2.56 O 4.81	1477	307
		4	.27 Ba- .10 S- 2.12 O 1.47	—	—
		5	1.04 Ba+2.12 S-10.4 O 4.93	587	119
MgO	(100)	1	Mg+O 1.17	10352	8800
		2	Mg+O 1.17	6258	5350
MgO	(111)	1	Mg-O .05	764	15300
		2	Mg+O 1.17	14175	12100
		3	Mg-O .05	132	2640
LiF	(111)	1	Li-F 1.19	10080	8470
		2	Li+F .01	~0	—
		3	Li-F 1.19	300	252
KBr	(111)	1	Br-K .21	1545	7360
		2	Br+K .91	2853	3140
KCl	(111)	1	Cl-K .78	2346	3010
		2	Cl+K 1.48	5620	3800
		3	Cl-K .78	334	428
		4	Cl+K 1.48	160	108
NaNO ₃	(211)	1	Na+N+ .88 O 1.96	29000	14800
		2	Na+N- .98 O .83	4171	5000
		3	Na+N+1.34 O 2.25	4148	1800
NaNO ₃	(111)	1	NO ₃ -Na 2.14	29400	13700
		2	NO ₃ +Na 3.26	16790	5150
		3	NO ₃ -Na 2.14	2243	1050
KI	(111)	1	K-I .01	85	—
		2	K+I .71	348	500
		3	K-I .01	24	—

found so far to the behavior of the majority of elements.

This type of data allows only a comparison of the signs of scattering lengths of different elements but not their absolute determination. The absolute determination of sign of scattering length will be discussed in Section 5. The over-all conclusions from Section 2 will be presented in Table VII. There, an attempt has been made to give the actual value of the scattering lengths for several elements. In calculating these values the attempt was made to obtain the best possible agreement of the observed intensities given in Table I, with the form factor after proper ac-

count was taken of the natural decrease of intensity with increasing order.

In x-ray analysis of crystals the Debye-Scherrer method of powder photography is extensively used. For neutrons, however, the intensity of the Debye-Scherrer maxima is rather small, and in most cases the method will be impractical. For example, in the case of microcrystalline graphite, we were able to detect only the maximum corresponding to the first reflection on the 001 plane.

A second possibility to study microcrystalline substances is to measure the total cross section of the substance for neutrons of wave-length

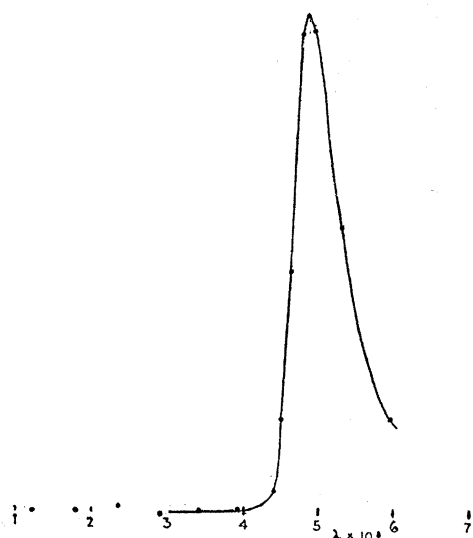


FIG. 3. Spectrum filtered through BeO.

close to $2d$ where d is the longest lattice spacing. An analysis of this case for substances containing two elements has been made by Fermi and Sachs, and indicates that the total cross section in this region depends strongly on whether the scattering lengths of the two elements have equal or opposite sign. The practicability of this method is somewhat limited by the fact that the theory applies only when the microcrystalline grain of the substance is exceedingly fine.

3. Spectrum of Filtered Neutrons

In the study of interference in gas molecules, somewhat simpler results are obtained with neutrons of energy far below thermal energies and which have a wave-length long compared with interatomic distances. Such neutrons are present in very small percentage in the Maxwell distribution at room temperature. Consequently, one cannot isolate such low energy neutrons by Bragg reflection because in the reflected beam there is a contamination by high orders which is many times more intensive than the first order. A simple way to avoid this difficulty, without use of extremely low temperature moderators, is to remove the high energy part of the Maxwell distribution by passing the beam of neutrons through a filter of a microcrystalline material of low absorption,² such as graphite or BeO. This

² Anderson, Fermi, and Marshall, Phys. Rev. **70**, 815 (1946).

filtering action of microcrystalline substances is due to the fact that neutrons of wave-length longer than $2d$, where d is the longest lattice spacing of the crystal, cannot satisfy the Bragg reflection condition for any of the many crystallites that they meet while crossing the filter. Consequently, they are scattered out of the beam only to a minimal extent. The production of very low energy neutrons by the filtering process has been previously reported² for the case of graphite, where the limiting wave-length is 6.7Å. In the present experiments BeO filters have been used, for which the limiting wave-length is 4.4Å. In order to get experimental proof of the correctness of the interpretation of the filtering process, we have investigated with the crystal spectrometer the spectrum of the neutrons filtered by a prism 40 cm long and containing 100 g/cm². The side dimensions of the prism were about 10×10 cm. The analysis was carried out with a crystal of celestite in the 001 plane. The results are plotted in Fig. 3, where the wave-lengths are plotted on the abscissae, and the observed intensities are the ordinates. The figure shows clearly that the spectrum of the filtered neutrons has negligible intensity for short wave-length, rises abruptly to a maximum value at wave-length of approximately 4.5Å, and after that, decreases gradually with increasing wave-length.

The decrease of intensity on the long wave-length side is somewhat more rapid than corresponds to the Maxwell distribution. This probably is due to the fact that the neutrons emerging from the graphite column are not fully slowed down to the lowest energies of the Maxwell distribution by the collisions in graphite.

The fact that microcrystalline BeO and Be metal are good filters for neutrons is an indication that the scattering lengths of Be for the two spin orientations of the neutron do not differ very considerably. Indeed if this were the case one would expect a strong incoherent scattering that should not vanish even for wave-lengths longer than $2d$. For filtered neutrons we found a residual cross section for BeO of 0.7×10^{-24} cm². If all this residual cross section were due to the difference in the scattering lengths a_1 and a_2 for the two spin orientations, it could be calculated that a_1 and a_2 certainly have the same sign and that their ratio is about 2. Probably the actual

values are appreciably closer, because part of the observed residual cross section is certainly due to crystal imperfections and to thermal agitation.

4. Scattering of Neutrons by Gas Molecules

Some conclusions on the scattering of neutrons can be drawn from the study of the total cross section of gas molecules. For neutrons of wave-length short compared with interatomic distances the molecular scattering cross section is the sum of the individual scattering cross sections of the atoms in the molecule. When neutrons of longer wave-length are used, a number of complications arise, partly due to interference effects, and partly to the fact that the scattering now is due to atoms that no longer can be considered free.

The theoretical calculation of the cross section has been carried out by Teller and Schwinger³ for the case of the H₂ molecule for low rotational states. For heavier molecules, such as N₂ or O₂, even when low temperatures and slow neutrons are used, a fairly large number of rotational states is always excited so that a detailed calculation of the contributions of all rotational states becomes impractical. Simple results can be calculated by neglecting the neutron mass in comparison with the mass of the atoms in the molecule. In this approximation one obtains results identical with those of the classical theory of interference. The approximation is quite good in the case of x-rays, where the particle scattered is a photon of very small effective mass, and has been used currently by Debye and his co-workers. For scattering of neutrons from molecules such as N₂, O₂, etc., the approximation is not nearly as good. The corrections, however, have not been calculated, and the results will be compared with those of the approximate theory except in the case of H₂ where the theory of Teller and Schwinger has been used.

The scattering cross sections of several molecules have been determined using very low energy neutrons obtained by filtration through a BeO filter. The spectrum of these neutrons is given in Fig. 3. Although these neutrons are not monochromatic they belong to a fairly narrow band of average wave-length 5.1Å. For the

measurements at room temperature the gas under investigation was contained in a long aluminum tube of 2.5" i.d. and 365 cm long. Pressures up to 2 atmospheres were used. On H₂ we performed some measurements at liquid air temperature and in this case a tube 24" long was used. A copper coil was soldered around this tube and liquid air was circulated in it. The entire assembly was protected with rock wool insulation. The conditions were such that the ortho-para ratio was practically the same as at room temperature.

Only in the cases of CF₄ and H₂ measurements were performed also for neutrons of shorter wave-lengths obtained by Bragg reflection on a fluorite crystal.

In Table II the total cross sections of BeO-filtered neutrons observed for various gases at room temperature are collected. In the third column of the table is given the sum of the total cross sections of the constituent atoms. One notices considerable differences between this sum and the observed cross sections. Except for the case of hydrogen which will be discussed later these differences are explained only to a small extent by the fact that the gas molecules are not at rest (so-called Doppler effect), and are due mostly to the interference of the waves scattered by the constituent atoms. In column 4 the values of σ calculated from the classical interference theory (see above) are given. For diatomic molecules containing two different atoms one obtains two different results, depending on the assumption that is made for the sign of the scattering lengths of the two atoms. In the cases of CO₂ and CF₄ a reasonably good agreement between theory and observation is obtained only if the scattering lengths of the component atoms are assumed to have the same sign. The same seems to be true for N₂O, though the sensitivity

TABLE II. Cross sections for $\lambda = 5.1\text{\AA}$
(gas at room temperature).

Molecule	σ (observed)	Sum of the total cross sections of constituent atoms	σ calculated from classical interference theory
CO ₂	24.5	13.0	24.8 or 4.1 for opposite phase
N ₂ O	57.8	34	55 or 41 for opposite phase
O ₂	16.2	8.2	13.2
N ₂	47.4	30	44.4
CF ₄	41.5	21	38 or 7.5 for opposite phase
H ₂	170	42	

³ Teller and Schwinger, Phys. Rev. 52, 286 (1937).

TABLE III. Cross section of CF_4 (room temperature).

$\lambda \times 10^8$	Velocity of neutrons (m/sec.)	$\sigma \times 10^{24}$
.881	4590	19.5
1.099	3600	19.0
1.316	3000	21.5
1.492	2650	20.8
5.1	775	41.5

to a change in relative sign of the scattering length is not so pronounced here.

The very large cross section observed for H_2 is strongly perturbed by the Doppler effect because here the thermal agitation velocity of the molecules is larger than the velocity of the neutrons, so that the phenomenon can be better described by saying that the molecules hit the neutron rather than the opposite. This fact is brought out by the strong temperature dependence of the cross section which drops from $170 \times 10^{-24} \text{ cm}^2$ to $81 \times 10^{-24} \text{ cm}^2$ when the H_2 is cooled from room temperature to liquid air temperature. This point will be discussed later.

Table III gives the cross sections observed for CF_4 at room temperature for neutrons of various velocities. The table shows a rise by about a factor 2 when the wave-length increases from 1.5 to 5.1A. Up to 1.5A the cross section is fairly close to $21 \times 10^{-24} \text{ cm}^2$, namely to the sum of the cross sections of one atom of carbon and four of fluorine. The rise is due in large measure to interference.

Tables IV and V give the results obtained on H_2 at room temperature and at liquid air temperature respectively. At room temperature one notices a steady rise in cross section with decreasing velocity of the neutrons. As previously indicated, this is to a large extent due to the thermal agitation velocity of the gas molecules.

TABLE IV. Cross section of H_2 at room temperature.

$\lambda \times 10^8$	Velocity (m/sec.)	$\sigma \times 10^{24}$	Factor for Maxwell distribution	σ/factor
.76	5212	50.9	1.048	48.6
.93	4256	53.2	1.071	49.7
1.11	3546	56.2	1.099	51.1
1.46	2714	61.2	1.172	52.2
1.68	2347	64.4	1.231	52.3
2.24	1765	68.0	1.407	48.3
5.1	775	169.7	2.514	67.5

A correction factor, calculated on the assumption that the cross section is constant, is given in the fourth column. The last column gives the cross section corrected by this factor.

The data of Table V taken at liquid air temperature can be most easily compared with the Teller-Schwinger theory because at this temperature practically only rotational states 0 and 1 are involved. Column 3 of this table gives the observed cross section and column 4 gives the cross section corrected for the thermal agitation velocity of the gas molecules. The fifth column gives the theoretical values calculated with the Teller-Schwinger theory, assuming $a_0 = -2.40 \times 10^{-12}$ and $a_1 = 0.54 \times 10^{-12}$ for the singlet and triplet state of the neutron proton system. The agreement is not too good especially for the lowest velocity, though the last value may be vitiated by the large Doppler correction.

By performing a similar set of measurements on deuterium one might be able to draw some conclusions of the spin dependence of the cross section, and we hope to make such measurements in the future.

5. Total Reflection of Neutrons of Mirrors

The total reflection of neutrons on mirrors is theoretically expected to occur at very small glancing angles for substances with a positive scattering length. The index of refraction is given by

$$n = 1 - \lambda^2 N a / 2\pi \quad (9)$$

where a is the scattering length taken with the proper sign, N is the density of atoms in the mirror, and λ is the wave-length. Consequently n is less than 1 (case of total reflection) if a is positive. The limiting glancing angle is then

TABLE V. Cross section of H_2 at 83°K (ortho-para ratio unchanged from room temperature).

$\lambda \times 10^8$	Velocity (m/sec.)	$\sigma_{\text{total}} \times 10^{24}$ observed	$\sigma_{\text{Sc}} \times 10^{24}$ corrected for absorption and Doppler effect	Teller-Schwinger theory
0.836	4730	44	43	—
1.202	3290	51	49	—
1.638	2420	55	52	56
1.884	2100	56	51	57
5.1	775	81	52	80

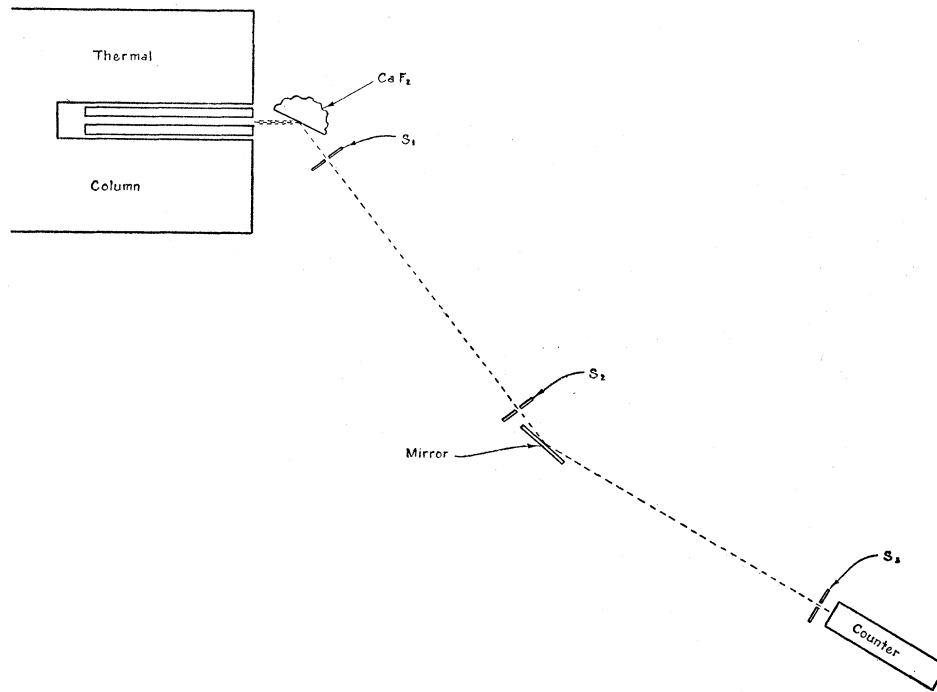


FIG. 4. Monochromatic total reflection on mirrors.

given with very good approximation by

$$\theta_0 = [2(1-n)]^{1/2} = \lambda(Na/\pi)^{1/2}. \quad (10)$$

In most cases $(n-1)$ is of the order of magnitude of 10^{-6} and the limiting angle of the order of $10'$. The total reflection of neutrons on mirrors has been previously observed⁴ using non-monochromatized thermal neutrons. In this case of course one cannot observe a limiting angle because θ_0 [see (10)] is proportional to λ so that neutrons of the various wave-lengths of the Maxwell dis-

tribution drop out of the reflected beam gradually as the glancing angle is increased. In spite of this, the fact that strong reflection is observed on a substance can be construed as good evidence that the index of refraction for that substance is less than one, and that therefore a is positive. This is so because, although a substance which is not totally reflecting has a finite reflection coefficient, it is so small that no very prominent reflection could be observed in such cases. From the quoted experiments it could therefore be

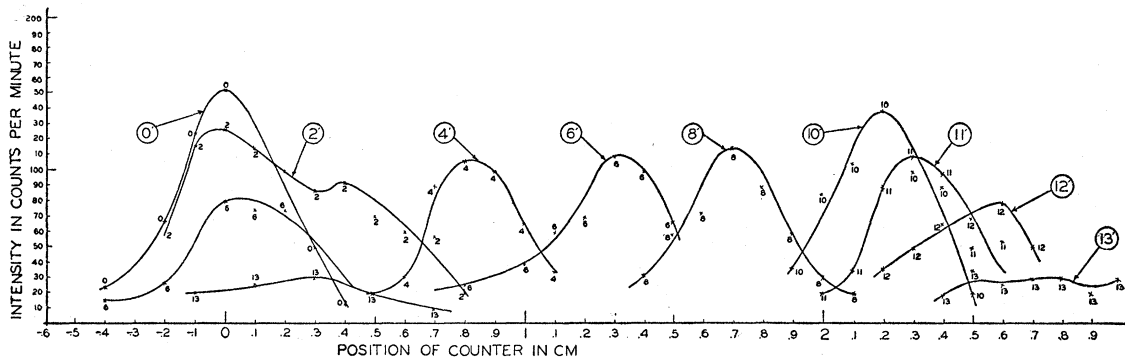


FIG. 5. Typical results of measurements of a reflection on a Be mirror.

⁴Fermi and Zinn, Phys. Rev. 70, 103 (1946).

TABLE VI. Limiting angle for total reflection of neutrons of 1.873A.

Mirror	Limiting angle (minutes)	
	Observed	Calculated
Be	12.0	11.1
C (graphite)	10.5	8.4
Fe	10.7	10.0
Ni	11.5	11.8
Zn	7.1	6.9
Cu	9.5	9.5

concluded that the following elements, Be, Cu, Zn, Ni, Fe, C, for which strong reflection has been observed, have positive scattering length.

In order to test conclusively the theory of total reflection, the reflection experiments were repeated using monochromatic neutrons. This made it possible to observe the sharp drop of the reflected intensity at the limiting angle and to measure this angle.

The experimental arrangement is shown schematically in Fig. 4. A collimated beam of thermal neutrons emerging from the thermal column is made monochromatic by Bragg reflection on a fluorite crystal. It is further collimated by passing through two very narrow cadmium slits, S_1 and S_2 , 2 mm wide and about 3.5 meters apart. The narrow monochromatic beam emerging from the second slit falls on the mirror which can be rotated by small angles. The reflected beam is detected by a BF_3 proportional counter about 3.5 meters away and with a cadmium entrance slit 2 mm wide. By moving the counter across the beam, both direct and reflected beam are detected.

Typical results of these measurements are plotted in Fig. 5 for the case of a reflection on a Be mirror. The abscissa is the position of the detector expressed in centimeters measured transversely to the direction of the beam. The ordinate is the neutron intensity in counts per minute. The curves whose experimental points are marked 0, 2, 4, etc., correspond to the intensity plots found when the mirror is set at an angle of 0, 2, 4, etc., minutes to the direction of the neutron beam. When the mirror is parallel to the beam, there is observed a single maximum corresponding to that of the main beam. When the mirror is rotated by 2' the reflected beam is barely resolved from the direct beam. At 4' the two maxima are well separated. For larger angles the direct beam has not been drawn in the plot

TABLE VII. Scattering lengths.

Element	$a \times 10^{12}$	$4\pi a_{\text{at}}^2 \times 10^{24}$	$\sigma_{\text{scat}} \times 10^{24}$	Remarks
Ba	0.79	7.7	8	C
Be	.89	8.0	6.1	M
Br	.56	3.9	<7	C
C	.67	4.8	4.8	A
Ca	.79	7.5	9.5	C
Cl	1.13	15	15	A, no spin and isotope effect considered
Cu	.81	8.0	7.2	M
F	.60	4.1	4.1	A, no spin effect considered
Fe	.82	8.1	9.2	M
H	-.39	.48	21	From theory
I	.36	1.6	1.6	A, no spin effect considered
K	.35	1.5	1.5	A, no spin and isotope effect considered
Li	-.59	3.4	—	C
Mg	.56	3.7	4	C
Mn	-.44	2.4	2.4	A, no spin effect considered
N	.87	8.3	8.3	A, no spin effect considered
Na	.56	3.7	3.5	C
Ni	1.09	14	13	M
O	.61	4.1	4.1	A
Pb	.48	2.9	10	C
S	.28	.95	1.1	C
Zn	.58	4.1	3.6	M

except for the largest angle, 13', at which the reflected beam has completely disappeared and also the direct beam has disappeared because the mirror has turned by such a large angle that it completely cuts off the main beam. These points therefore correspond to the background, which actually was about 20 counts per minute.

By inspecting the plot one sees that the intensity of the reflected beam drops sharply to background between the positions 10 and 13 minutes. This drop is not quite sudden because the angular resolution was about two minutes. A study of the intensity shows the limiting angle to be 12 minutes.

In Table VI the values of the limiting angles of several mirrors measured for neutrons of de Broglie wave-length $\lambda = 1.873\text{A}$ are given. The experimental values given in the second column of the table should be compared with the theoretical values calculated from formula (10) given in the third column. The agreement is excellent except in the case of carbon, for which a not too good graphite mirror was used.

The absolute determination of the sign of the scattering length for the totally reflecting elements, taken together with the results of Sections 2 and 4, allows one to determine the actual sign for a large group of elements.

6. Conclusions

In Table VII all the data available at present on scattering length have been summarized. The

absolute sign given in the table has been obtained primarily from the data of Section 5 on total reflection, and indirectly, by combining these data with those of Section 2 on intensities of Bragg reflections, and Section 4, on molecular scattering. The absolute values have been obtained in various ways.

For the elements having only one isotope or one strongly dominant isotope, and no nuclear spin, the absolute value of the scattering length can be calculated from formulae (5) and (2) provided the total scattering cross section is known. For example, this method has been applied in assigning the value 0.61×10^{-12} cm for the scattering length of oxygen in a crystal. This differs by a factor 16/17 from the scattering length 0.57×10^{-12} cm for a free oxygen atom. In some cases this same method has been applied to elements having several important isotopes, or a spin, or both. In such cases the absolute value is naturally much less reliable. The method has been used when no better way was available. In the last column of Table 7 the code letter *A* has been used to denote values obtained by this method.

The results of Section 2 allow the scattering lengths of several elements to be related directly or indirectly to that of oxygen whose scattering length 0.61×10^{-12} has been used as a standard for these cases.

For the five elements Na, K, Cl, Br and I, which are not conclusively related to the others (see Section 2), Cl has been taken as a standard with a scattering length of 1.13×10^{-12} cm. The sign has been taken positive partly on account of the evidence from Bragg reflection on NaNO_3 , as discussed in Section 2. Furthermore, both theoretical and experimental evidence indicate that cases of negative scattering length are very improbable. The probability that five elements all having the same sign should be negative is therefore exceedingly small.

In other cases the value of the scattering length has been calculated from the value of the limiting angle using formula (10). Code letter *M* indicates these instances.

In the third column of Table VII is given the expression $4\pi a^2(A/A+1)^2 \times 10^{24}$. If there are no

important isotopes and no spin this should coincide with the scattering cross section. Otherwise it should in general be smaller, the more so the greater the differences are in the scattering lengths for the various isotopes and spin orientations.

The values in column 3 therefore should be compared with the experimental values of the scattering cross section given in column 4. Naturally the comparison is trivial for the cases designated by *A*, where formulae (2) and (5) have been used to calculate *a*. Comparison in the remaining cases shows that usually the data of the third and fourth columns are rather close. The few exceptions for which the data in column 3 are less than those of column 4 can be explained as due to experimental inaccuracy. The fact that they are close seems to indicate that, in general, the spin and isotope dependence of the scattering length is not extremely pronounced.

There are two notable exceptions. One is hydrogen, a case well known from the Teller-Schwinger theories. The other one is lead where perhaps some of the many isotopes may have a negative scattering length.

The table shows that the scattering length is positive in the great majority of cases as discussed in Section 1.

This work was performed at the Argonne National Laboratory, and we greatly benefited by the help of the staff of the laboratory in providing and operating the neutron source, and in supplying many of the experimental facilities needed. Several of the crystals used were loaned to us by the Field Museum in Chicago through the courtesy of Mr. Changnon, by Dr. Howland of the Mineralogy Department of Northwestern University, and by Dr. Fisher of the Geology Department of the University of Chicago. Also some specimens were obtained from the Union Carbide and Carbon Research Laboratories, Inc., and the Eagle Picher Lead Company. Mr. Warren Nyer helped in some of the experiments.

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