lack of separation of the angular distribution into $\cos^2\theta$, $\cos^4\theta$, etc., terms in the high energy experiments.

A measurement of the value of B, the coefficient for the $\cos^4\theta$ asymmetry, would have the greatest significance for determining the magnitude of the spin-orbit coupling in the nuclear interactions. It would determine just how large the spin-orbit coupling must be made to account for results such as those of Fig. 2.

The authors are grateful to Mr. Geoffrey Chew for helpful calculations in connection with this work.

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The Diffraction of Neutrons by Crystalline Powders

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The powdered crystal technique, which has been shown for the case of x-rays to be best suited for accurate, integrated intensity measurements, has been applied to neutron diffraction. Neutrons from the Clinton pile are monochromatized by reflection from a single crystal and the diffraction patterns produced when these neutrons fall on specimens of crystalline powders have been studied. These studies have given (a) a check on some aspects of the diffraction theory, (b) the magnitude and sign of the scattering amplitudes from various nuclear species from which information is obtained on the spin dependence of the scattering, which has a bearing on the magnitude and range of nuclear forces, and (c) an improvement in the techniques and a better understanding of the problems involved so that results can be more readily obtained on the diffraction by other crystals, such as those containing hydrogen or deuterium.

Diffraction measurements have been obtained on diamond, graphite, Al, Na, NaBr, NaCl, and NaF in which all intensity measurements were standardized against diamond to which a definite cross section was assigned on the basis of total cross-section measurements. The scattering of carbon, Al, and F was found to have no measurable spin dependence. The scattering by Na, however, shows a considerable spin dependence as evidenced by a Bragg scattering cross section of 1.51 barns as against a total scattering cross section of 3.7 barns. Measurements have been made on a number of other crystals with the purpose of determining the phase of scattering. A table showing the scattering phase for a number of elements is given.

1. INTRODUCTION

EXPERIMENTAL work on the diffraction of neutrons by crystals, directed towards obtaining information about the diffraction process and its dependence on the crystal and nuclear properties of various substances, was started at Clinton Laboratories in 1945 by E. O. Wollan and R. B. Sawyer. The first measurements along this line were made with single crystals; the results gave information regarding the phase of nuclear scattering. It was found, however, that with single crystals it would be difficult to make measurements of the diffracted intensity with sufficient accuracy to permit reliable conclusions to be drawn regarding the effect on the intensity of various factors such as nuclear spin, presence of more than one isotope, characteristic temperature of the crystals, crystal structure, etc.

By using the powdered crystal method one can almost completely eliminate the effects of extinction and of crystal distortion and, as has been shown in the case of x-rays, accurate intensity measurements can be made if a sufficiently intense source of monochromatic radiation is available. The flux from the Clinton pile was found to be just sufficient to permit one to use the powder diffraction method. However, since no monochromatic lines are available with neutron sources, it is necessary to first reflect the neutrons from a single crystal (or use a mechanical velocity selector) to obtain a monoenergetic beam and this fact also increases the difficulty of obtaining sufficient intensity for the application of this method. The low intensity encountered in this work has made progress less rapid than would be desired, but recent changes in the apparatus have gone far in the direction of improving the situation.

The recently published experimental work of Fermi and Marshall[1] and Fermi, Sturm, and Sachs[2] performed at the Argonne National Laboratory have been directed along the same lines as the present research. Fermi and Marshall have studied Bragg reflections from a variety of single crystals, thereby obtaining information on the relative phase of scattering and the coherent scattering cross sections for various elements. On the other hand, Fermi, Sturm, and Sachs have observed the variation of total scattering cross section by crystalline powders as a function of neutron energy. For materials possessing a favorably large ratio of scattering to capture cross section, pronounced changes in the crystal scattering cross section as a function of changing neutron wave-length should result from crystal diffraction effects as Halpern, Hamermesh, and Johnson[3] have predicted. The information available from the present diffraction experiments duplicates in many respects that from the above procedures.

2. EXPERIMENTAL ARRANGEMENT

The data which are being reported here were taken during the past year with an experimental arrangement which was accepted at the time as being satisfactory. The long hours of readings which were required to produce data with good statistical accuracy, however, led recently to the incorporation of improvements in the technique which have materially increased the efficiency of the work, and all the data have been retaken with the present experimental arrangement.

The arrangement of the apparatus, including the monochromator, the spectrometer for holding the powder specimens, the counter and the associated shielding, is shown in Fig. 1. A beam of neutrons emerges from the pile through a 1 in. ×1 in. square hole in a one-foot long shielding block. The neutrons emerging from the pile through this hole have a maximum angular divergence of about one degree. The monochromating crystal (NaCl), on which the neutrons fall, is located on a turntable attached to the pile face, and the region around the crystal is heavily shielded with borated paraffin and lead. In our earlier work the cleavage face (200 plane) of the NaCl crystal was used as the reflecting surface. The face of the crystal has now been cut so that it makes an angle of 6° with the (200) reflecting planes and this permits the primary neutron beam of 1-in. width to give a diffracted beam of $\frac{3}{8}$-in. width. The situation can be understood from the enlarged drawing of the crystal shown in Fig. 1. This method of cutting a crystal to permit the use of a wide primary beam was first suggested for x-ray work by Fankuchen.[4] The method has proved effective with neutrons, our beam strength having been improved by a factor of about two. The monochromatic beam from this crystal passes out through the shield through a long boron carbide lined slit, 1 in. high by $\frac{3}{8}$ in. wide, and passes over the center of a spectrometer table on which the powder crystal specimens are held.

The crystal powders are contained in flat cells with thin aluminum windows (which give negligible scattering) and these cells are held on the spectrometer table in the conventional position for diffraction by transmission. The counter has an active length of 18 inches and is filled with BF_3 gas in which the boron is enriched in the B^{10} isotope. The counter is shielded with four inches of paraffin and $\frac{1}{2}$ in. of B_4C. It is arranged to rotate about the center of the spectrometer table; the heavy load caused by shielding, is supported by cables leading to a bearing above the spectrometer table in line with the axis of rotation.

The process of taking data is now on an automatic basis. The electrical impulses from the proportional counter are fed to an amplifier and scaler and to a printing recorder. The counter

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[1] E. Fermi and L. Marshall, Phys. Rev. **71**, 666 (1947).

[2] E. Fermi, W. J. Sturm, and R. G. Sachs, Phys. Rev. **71**, 589 (1947).

[3] O. Halpern, M. Hamermesh, and M. H. Johnson, Phys. Rev. **59**, 981 (1941).

[4] I. Fankuchen, Nature **139**, 193 (1937).

and the powder specimen are rotated by a motor at a low angular velocity (the counter at 15′ and the specimen at $7\frac{1}{2}'$ of arc per four minutes of time) and at four-minute time intervals the printing recorder stamps the number accumulated on a mechanical counter and then resets the counter to zero. Alternate four-minute intervals are taken with and without cadmium in the beam, this being accomplished automatically with a solenoid. Adjustable microswitches for reversing the motor drive can be set to keep the spectrometer operating within any desired angular range. The advantage of automatic operation cannot be overestimated since the best data can be taken during the evening and night shifts.

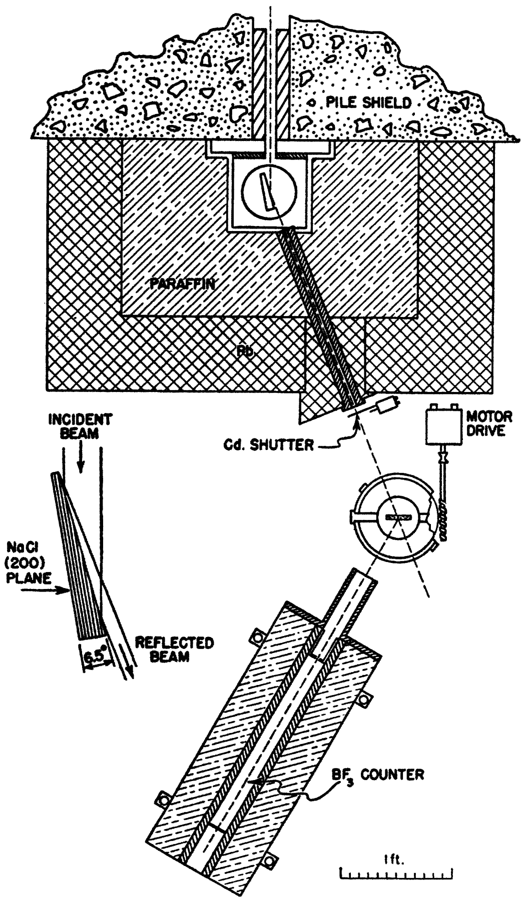

FIG. 1. Arrangement of apparatus, showing the monochromating crystal (detailed in left center) collimating slits, shielding, second spectrometer with location of powder specimen and counter.

3. ANALYSIS OF RADIATION FROM FIRST CRYSTAL

In the study of the diffuse scattering from crystals it is important to know the energy distribution of the neutrons incident on the crystals or crystal powders being investigated.

The quality of the neutron beam reflected from the first crystal depends on the energy spectrum, the angular divergence of the neutrons falling on the crystal, and on the perfection of and the amount of diffuse scattering from the monochromating crystal.

To analyze this beam we have made rocking curves on both sides of the beam with a second crystal set in the transmission position on the spectrometer table. When the reflecting planes of the first and second crystal are parallel, one obtains a narrow rocking curve (width at half-maximum = 10 min. of arc) with a high peak intensity. This rocking curve is a measure of the imperfection of the two crystals and its width is practically independent of the energy in the incident beam for the neutrons which have been Bragg reflected from the first crystal. On the other hand, the rocking curve, taken in the non-parallel position, is a measure of the energy spread in the neutron beam falling on it. The energy distribution in the beam can then be obtained from the shape of the intensity distribution in the non-parallel position. A plot of the energy distribution thus obtained is shown in Fig. 2 without correction for crystal imperfection which, if made, would narrow the curve by about 10 percent.

The amount of radiation diffusely scattered from the first crystal was determined by measuring the intensity through the slit when the first crystal was set at an angle of 2° off the maximum of the Bragg peak. In this position the measured intensity was found to be about 3 percent of that obtained with the monochromator in the optimum position for Bragg reflection.

A further test of the quality of the radiation from the monochromating crystal was made by taking rocking curves with the second crystal at various angles both inside and outside of the first-order peak. At no other angular position was there evidence of radiation outside of the energy range shown in Fig. 2, except that corresponding to second-order reflection of radiation of $\frac{1}{2}\lambda_0$, $(4E_0)$, where λ_0, (E_0) are the wave-length and energy of the main first-order reflection from the monochromating crystal. The strength of this second-order contamination as determined from a rocking curve at $\sin\theta = \frac{1}{2}\sin\theta_0$ was found to be 1.6 percent of the first-order peak.

We feel that the above analysis shows our monochromatic beam to be of sufficient purity to introduce no appreciable errors in our diffraction measurements.

4. THEORETICAL CONSIDERATIONS

Although the scattering of slow neutrons and the scattering of x-rays are in many respects very similar problems, there are some distinct differences which make the scattering of slow neutrons a field in itself.

With x-rays, the atomic electrons are the scattering entities and the cross section for scattering by a single electron can be accurately calculated. On the basis of classical theory one obtains the well-known Thomson formula which is valid for bound electrons. A free electron, however, takes up momentum from the incident photon and in this case one must apply quantum theory which leads to the Klein-Nishina Compton scattering formula. The scattering by an atom is obtained by summing the scattering amplitudes of all the electrons with respect to their relative positions in the atom and this gives the atomic scattering factor which is defined in terms of the relative scattering by the whole atom to that of a single bound electron. With x-rays these atomic scattering factors can be determined experimentally, and they can also be approximately determined theoretically by the method of self-consistent fields.

With neutrons the scattering is almost entirely nuclear and, since so little is known about nuclear structure, it cannot be expected that scattering cross sections, in general, can be determined theoretically. Aside from this fundamentally different mechanism of scattering, there exist a number of other basic differences between neutron and x-ray diffraction which will be of interest in connection with the experimental data and a general review of these is given in the following sections. Details on these discussion points can be found in earlier papers.[3, 5–14]

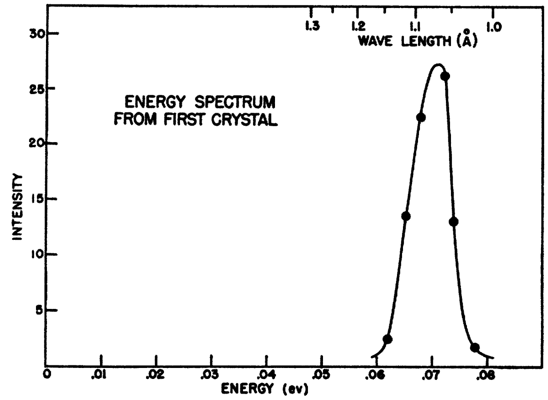

FIG. 2. Energy and wave-length distribution of neutrons reflected from monochromating crystal.

5. SCATTERING THEORY

The theory of the scattering of neutrons by nuclei gives a relation between the scattering amplitude and the nuclear potential. For the case of slow neutrons the theory is simplified by the fact that there is only *s* scattering which is spherically symmetric.

The wave equation representing the interaction of a neutron with a nucleus is

$$\nabla^2\psi + k'^2\psi = 0 \qquad (1)$$

where

$$k' = \left(\frac{2m'(E-V)}{\hbar^2}\right)^{\frac{1}{2}}$$

is the wave number inside the nucleus whose potential function is $V(r)$ and m' is the reduced mass of the system. Correspondingly, the incident wave is represented by the same equation with $V=0$ and then $k = (2mE/\hbar^2)^{\frac{1}{2}} = 2\pi/\lambda$, where λ is the wave-length of the incident neutrons and m is the neutron mass.

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[5] E. Fermi, Ricerca Scient. **7**, 13 (1936).

[6] G. C. Wick, Physik. Zeits. **38**, 403 (1937).

[7] J. Schwinger and E. Teller, Phys. Rev. **52**, 286 (1937).

[8] H. A. Bethe, Rev. Mod. Phys. **9**, 69 (1937).

[9] I. Pomeranchuk, Physik. Zeits. Sowjetunion **13**, 65 (1938).

[10] R. J. Seeger and E. Teller, Phys. Rev. **62**, 37 (1942).

[11] R. Weinstock, Phys. Rev. **65**, 1 (1944).

[12] H. Feshbach, D. C. Peaslee, and V. F. Weisskopf, Phys. Rev. **71**, 145 (1947).

[13] M. L. Goldberger and F. Seitz, Phys. Rev. **71**, 294 (1947).

[14] R. J. Finkelstein, Phys. Rev. **72**, 907 (1947).

TABLE I. Scattering data for diamond and graphite.

hkl	P_{hkl} (integrated counts/min.)	e^{2W}	$k^2f_0^2$	f_0 (10^{-12} cm)	$\sigma_B=4\pi f_0^2$ (barns)	σ (barns)
			Diamond			
111	253	1.018	10.5	0.64	5.2	5.2*
220	272	1.048	9.5			
311	216	1.067	9.7			
331	151	1.119	9.9			
			Graphite			
002	135		10.6	0.66	5.5	

* From our transmission measurements.

The solution of the wave equation[15] at large distances r from the nucleus (neglecting absorption) can be represented as the sum of an incident plane wave (along z) and a radially scattered wave

$$\psi=e^{ik'z}+fe^{ikr}/r, \tag{2}$$

where f is the complex amplitude of the scattered wave.

It can be readily shown that f depends on the phase shift of the total wave with respect to the incident wave and this phase shift depends on the nuclear potential $V(r)$. For slow neutrons there is only s scattering and, in this case, only the spherically symmetric part of the solution need be considered. The spherically symmetric part of the incident plane wave can be represented as the sum of an incoming and an outgoing wave

$$(\sin kr)/kr=(e^{ikr}-e^{-ikr})/2ikr. \tag{3}$$

The corresponding spherically symmetric part of the total wave with the nuclear potential turned on will be given by the same function with different phase,

$$\frac{A\sin(kr+\eta_0)}{kr}=\frac{Ae^{i(kr+\eta_0)}-Ae^{-i(kr+\eta_0)}}{2ikr}. \tag{4}$$

The difference between Eqs. (3) and (4) will then represent the scattered wave

$$f\frac{e^{ikr}}{r}=\frac{(Ae^{i\eta_0}-1)e^{ikr}-(Ae^{-i\eta_0}-1)e^{-ikr}}{2ikr}. \tag{5}$$

In the scattered wave, however, there must be no incoming wave and for this condition to be satisfied, $A=e^{i\eta_0}$. Substituting this value for A back into Eq. (5) gives

$$f=(e^{2i\eta_0}-1)/2ik, \tag{6}$$

and the cross section for scattering will be

$$\sigma_{\text{scat}}=4\pi|f|^2=4\pi\sin^2\eta_0/k^2. \tag{7}$$

The phase shift η_0 is very small except in the near vicinity of a resonance level and hence in most cases only the first power of η_0 in the ex-

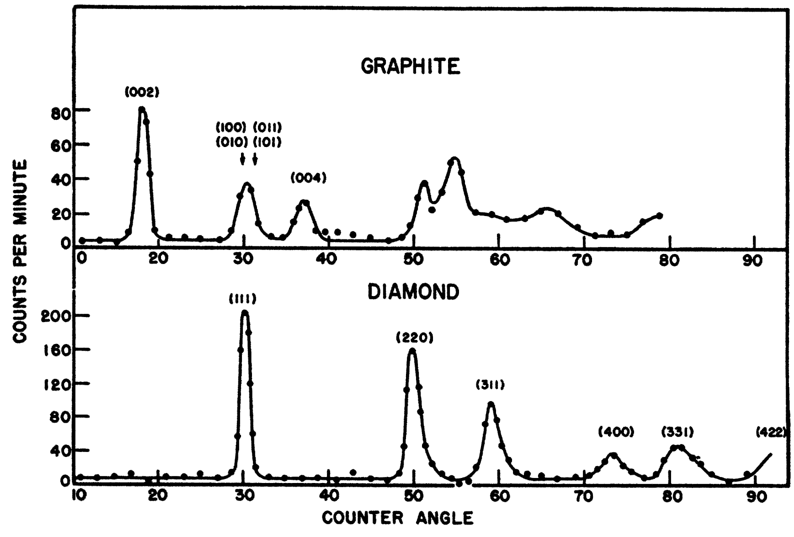

FIG. 3. Powder diffraction patterns for diamond and graphite. The major part of the diffuse scattering in these patterns arises from multiple scattering in the samples.

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[15] See for example N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, 1933).

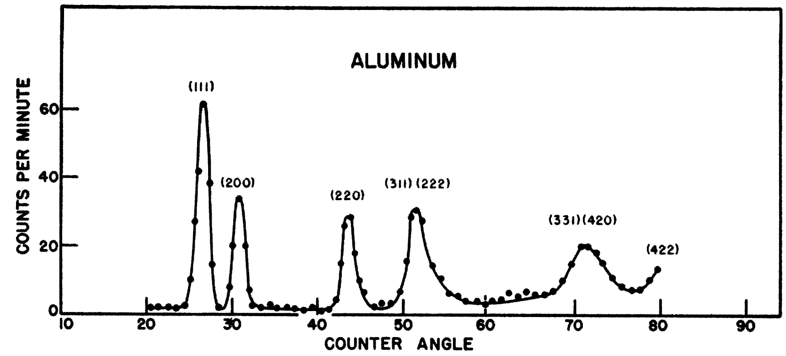

FIG. 4. Powder diffraction pattern for aluminum. The diffuse scattering at small angles arises primarily from multiple scattering. At the larger angles it is mostly temperature diffuse scattering.

pansion of Eq. (7) need be considered so that

$$f=\eta_0/k, \tag{8}$$

and

$$\sigma=4\pi\eta_0^2/k^2. \tag{9}$$

Positive or negative phase shifts correspond to positive or negative scattering amplitudes and, hence, the corresponding scattered waves for these two cases will be 180° out of phase with each other. Positive phase shifts occur only in the vicinity of a resonance and, hence, they will be less common than negative phase shifts.

The measurement of phase shifts or scattering amplitudes will constitute a good check on any theory of nuclear structure, since, for such a theory to be satisfactory it must be capable of yielding these quantities.

For hydrogen the scattering cross section can be evaluated in terms of the binding energy and the range of forces in the triplet and singlet state of the deuteron. With heavy elements for which the nucleus behaves in most cases as an impenetrable sphere, rough estimates of the cross section can be made by taking $\sigma=4\pi a^2$ where the radius a is approximately equal to the nuclear radius as obtained from other sources of information.

6. BINDING, ISOTOPE, AND SPIN EFFECTS

The scattering cross section for a given nucleus depends on whether the nucleus is in a free or a bound state. The cross section for scattering by a rigidly bound nucleus will be greater than for the same nucleus in a free state by the square of the ratio of the neutron mass to the reduced mass for the neutron and the scattering nucleus, i.e., $[(A+1)/A]^2$ where A is the mass number of the scattering nucleus. Between the extremes of completely free scattering centers and completely bound centers there will be a region which may be loosely referred to as that of partial binding for which the reduced mass effect will lie somewhere between $[(A+1)/A]^2$ and 1. The coherent scattering by crystals will always correspond to rigidly bound scattering centers.

When scattering measurements are made on an element containing more than one isotope the results will correspond to an average of the scattering by these isotopes. If the measurements are made with free nuclei, the total scattering cross section averaged over the isotopes will be

$$\sigma_f=4\pi(p_1a_1^2+p_2a_2^2+\cdots), \tag{10}$$

where the p's are the isotopic abundances and the a's are the free nuclear scattering amplitudes for the respective isotopes. If the atoms are bound but still act independently as far as interference effects are concerned, the corresponding cross section will be

$$\sigma_{\text{bound}}=\sigma_f[(A+1)/A]^2$$
$$=4\pi(p_1f_1^2+p_2f_2^2+\cdots), \tag{11}$$

where the f's are the scattering amplitudes for bound scattering centers.

The coherent scattering by the various isotopes of an element randomly distributed in a crystal will be

$$\sigma_{\text{coh}}=4\pi(p_1f_1+p_2f_2+\cdots)^2, \tag{12}$$

since here the isotopic amplitudes must be combined algebraically into a total amplitude before squaring to obtain the intensity.

A situation similar to that of the isotope effect arises from the fact that neutron scattering may depend on the relative orientation of the spins

TABLE II. Scattering data for aluminum.

hkl	P_{hkl} (integrated counts/min.)	e^{2W}	f_0 (10^{-12} cm)	$\sigma_B=4\pi f_0^2$ (barns)	σ (barns)
111	82.0	1.08	0.330	1.38	
200	47.5	1.11	0.338	1.43	
220	20.3	1.23	0.334	1.40	1.48*
311 222	91.2	1.35 1.37	0.350	1.54	
331 420	73.9	1.67 1.71	0.345	1.50	
		averages	0.340	1.46	

* σ_{free} taken from literature as 1.37 barns.

of the neutron and the scattering nucleus. There will then be two amplitudes associated with each nucleus, one for parallel and one for antiparallel spins. The measured scattering cross section for a given nuclear species will then correspond to an average, weighted with respect to the probability for the two spin states. For scattering by free nuclei then

$$\sigma_f=4\pi\left[\frac{i+1}{2i+1}a_{i+\frac{1}{2}}{}^2+\frac{i}{2i+1}a_{i-\frac{1}{2}}{}^2\right], \tag{13}$$

where i is the spin of the scattering nucleus and the a's are the free scattering amplitudes for parallel and antiparallel spin. For the coherent scattering by bound nuclei

$$\sigma_{\text{coh}}=4\pi\left[\frac{i+1}{2i+1}f_{i+\frac{1}{2}}+\frac{i}{2i+1}f_{i-\frac{1}{2}}\right]^2. \tag{14}$$

These equations apply to the case where the nuclear spins are randomly oriented. In the case of ortho- and parahydrogen the situation is different since the spins of the two nuclei in the molecule have a definite orientation.

7. THE SCATTERING OF NEUTRONS BY CRYSTALS

The equations describing the intensity pattern of neutron diffraction by a crystalline powder are exactly similar to those characteristic of x-ray diffraction except for the absence of the polarization factor in the present case. As mentioned earlier, the present data have been taken with a flat plate of crystalline powder arranged in the transmission orientation and for this case, the power P_{hkl} diffracted in a selected Bragg peak is given by

TABLE III. Scattering data for sodium containing crystals.

Crystal	hkl	P_{hkl} (integrated counts/min.)	f_0 (10^{-12} cm)	f_0 (10^{-12} cm)	$4\pi f_0^2$ (barns)	$4\pi f_0^2$ (barns)	σ^* (barns)	σ^* (barns)
			Na	X	Na	X	Na	X
Na	110	11.3	0.345		1.50		3.7	
NaBr	111 200	10.2 48.5	0.335	Br 0.65	1.41	Br 5.3		Br 7.5
NaCl	111 200	30.5 72.5	0.358	Cl 1.01	1.61	Cl 12.8		Cl 15
NaF	111 200	13.7 88.5	0.348	F 0.65	1.52	F 5.2		F 4.5
		averages	0.346		1.51			

* Literature values $\times[(A+1)/A]^2$.

$$\frac{P_{hkl}}{P_0}=\frac{\lambda^3 l}{4\pi r}\,\frac{h\rho'}{\rho}\,\frac{e^{-\mu h\sec\theta}}{\sin^2 2\theta}\,j_{hkl}N^2F_{hkl}{}^2, \tag{15}$$

where P_0 is the power in the primary beam, λ is the wave-length of the neutrons, l is the height of the slit in front of the counter, r is the distance from sample to counter slit, ρ' is the density of the powder, ρ is the density of the solid crystal, $e^{-\mu h}$ is the transmission of specimen of thickness, h, θ is the glancing angle of reflection, j_{hkl} is the multiplicity factor, N is the number of unit cells per cm^3 of crystal, and F_{hkl} is the Bragg scattering amplitude per unit solid angle per unit cell of the crystal. F_{hkl} corresponds to the usual crystal structure factor in x-rays, except that here it is a true amplitude with the dimension of length. It is a linear combination of effective nuclear scattering amplitudes which we will designate by f_T, the subscript T being used to designate the amplitude obtained from direct measurement of a Bragg peak for a crystal at a given temperature T. For neutrons, as for x-rays, the loss in Bragg scattering due to the interaction of the neutrons with the lattice vibrations can be corrected for by the Debye-Waller temperature factor as Weinstock[11] has shown, and the true nuclear scattering amplitude f_0 for a given nucleus will be

$$f_0=f_Te^W,$$

where

$$W=M\frac{\sin^2\theta}{\lambda^2},\quad M=\frac{6h^2}{mk\Theta}\left(\frac{\varphi(x)}{x}+\frac{1}{4}\right).\tag{16}$$

$x=\Theta/T$, $\varphi(x)$ is the Debye function and Θ is the Debye characteristic temperature.

If the crystal specimen is monatomic, measurement of the absolute intensity of a single peak, plus a knowledge of the characteristic temperature of the crystal will suffice for the determination of f_0^2. With polyatomic crystals, two or more peaks must be measured to obtain values of f_0^2 for the individual atoms and even then the assignment of f_1^2 and f_2^2 to the nuclei 1 and 2 is only possible if it is known which of these cross sections is the larger. This may be determined without previous information by making measurements with a given element in several compounds. In x-rays this difficulty never arises since atomic scattering factors are known to increase with the atomic number.

In addition to the Bragg scattering, it is important to consider also the intensity of the neutrons diffusely scattered by a crystal. With x-rays the diffuse scattering consists of the Compton inelastic scattering and the temperature diffuse scattering some of which may also be inelastic. According to the Debye theory, the temperature diffuse scattering is given for a single crystal by $(1-e^{-2W})f_0^2$, where f_0 is the atomic structure factor. This theory has been extended by Zachariasen[16] to include the quantized interaction of the radiation with the Debye waves of the crystal. For the neutron case the corresponding theory has been developed by Weinstock[11] and extended by Seitz and Goldberger[13] and by Finkelstein.[14]

In the case of neutrons there will also be diffuse scattering arising from the spin and isotope effects. The over-all scattering by a crystal will then be made up of the coherent Bragg scattering plus the diffuse scattering arising from various sources. The over-all scattering cross section for a crystal as measured, for example, by a transmission experiment with correction for the effects of absorption, can then be written as

$$\sigma_{\text{crystal}}=\sigma_{\text{Bragg}}+\sigma_{DS}+\sigma_{DI}+\sigma_{DT},\tag{17}$$

where σ_{Bragg} is the total scattering in the Bragg peaks and σ_{DS}, σ_{DI}, and σ_{DT} are the diffuse scattering cross sections for the spin, isotope, and temperature effects, respectively.

As an example of the diffuse scattering arising from the spin effect, consider a crystal containing an element of one isotope ($\sigma_{DI}=0$) and having a high characteristic temperature ($\sigma_{DT}\sim 0$) and if, in addition, A is large, then $[(A+1)/A]^2\sim 1$ and we obtain

$$\begin{aligned}\sigma_{DS}&=\sigma-\sigma_{\text{Bragg}}\\&=4\pi\left[\frac{i+1}{2i+1}f_{i+\frac{1}{2}}^2+\frac{i}{2i+1}f_{i-\frac{1}{2}}^2\right]\\&\qquad-4\pi\left[\frac{i+1}{2i+1}f_{i+\frac{1}{2}}+\frac{i}{2i+1}f_{i-\frac{1}{2}}\right]^2\\&=4\pi\frac{i(i+1)}{(2i+1)^2}[f_{i+\frac{1}{2}}-f_{i-\frac{1}{2}}]^2.\end{aligned}\tag{18}$$

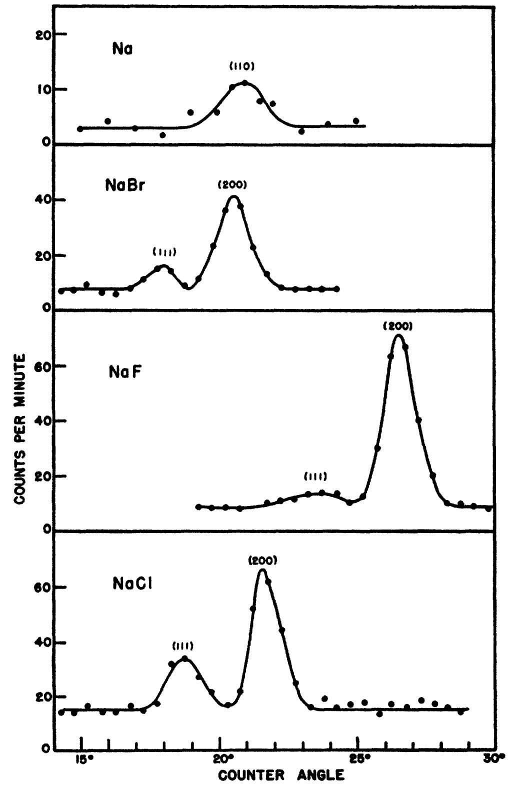

FIG. 5. Powder diffraction patterns for four Na containing crystals. The fact that the (111) peaks are smaller than the (200) peaks shows that Na, Br, F, and Cl all scatter with the same phase.

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[16] W. H. Zachariasen, *Theory of X-Ray Diffraction in Crystals* (John Wiley and Sons, Inc., New York, 1945).

TABLE IV. Scattering amplitudes for two spin states of sodium.

	$f_{i+\frac{1}{2}}$	$f_{i-\frac{1}{2}}$	
either	$+0.88$	$+0.02$	$\times 10^{-12}$ cm
or	-0.20	$+0.67$	

TABLE V. Sign of scattering amplitudes for various elements.

Positive scattering amplitude			Negative scattering amplitude
O	Pb	Na	Mn
Fe	N	K	Li
Mg	C	Cl	H
Ba	Be	Br	
Ca	Al	I	
S	Sr	D	
F			

It will be seen from this equation that the diffuse scattering due to the spin dependence will be zero when the scattering amplitudes for parallel and antiparallel spins are equal. This will be the case for a nucleus with zero spin. The other extreme is reached when the two weighted spin amplitudes are equal in magnitude but opposite in sign in which case there will be no Bragg peaks from a crystal, all the scattering will be diffuse.

For a crystal containing a light element, such as hydrogen, the spin diffuse scattering (for our wave-length $\lambda \sim 1$A) will be considerably reduced by the lack of complete binding (see following paper).

A situation similar to the one we have discussed here for the spin dependence will also exist with respect to the scattering by an element containing more than one isotope.

8. EXPERIMENTAL RESULTS

Diamond

The neutron diffraction measurements for diamond (800-mesh powder) are shown by the lower curve of Fig. 3. The measurements correspond to the difference in counting rates with and without a cadmium shutter in the beam incident on the specimen, the incident neutrons being monochromatic, as we have already pointed out, and of wave-length $\lambda = 1.057$A.

From the integrated intensities of the Bragg peaks one can determine values of F_{hkl}^2 from Eq. (16) except for a constant, k^2, which depends on the effective incident neutron intensity which it has not yet been possible to measure directly. From the measured values of $k^2F_{hkl}^2$ one can determine $k^2f_T^2$, where f_T is the effective Bragg scattering amplitude of the carbon atoms at the crystal temperature at which these measurements are made.

By applying the usual Debye-Waller temperature factor e^{2W} (Eq. (16)) to the measured values of $k^2f_T^2$ the effects of the lattice vibrations are eliminated and one obtains numbers proportional to the true Bragg scattering amplitudes for the carbon atoms in the crystal. The values of $k^2f_0^2 = k^2f_T^2e^{2W}$ are listed in Table I for four diamond peaks.

Whether or not k^2f^2 is proportional to the total nuclear scattering cross section of carbon can be determined by measuring both the Bragg scattering and the diffuse scattering by the crystal. If there are no spin or isotope effects with diamond, then one would expect no diffuse scattering other than that arising from the effects of lattice vibrations. Since C^{12} has zero spin, any spin diffuse scattering would have to be associated with C^{13}. Since this isotope is present in normal carbon to the extent of only 1.1 percent, one would expect its contribution to the total scattering to be very small.

The actual measured diffuse background from diamond, as shown in Fig. 4, is, however, quite appreciable even at small angles where the temperature diffuse scattering should be immeasurably small. This extra diffuse background can, however, be accounted for on the basis of multiple scattering by the sample. Although the attenuation of the neutron beam in traversing the sample was in this case only 20 percent, the fact that there is no true absorption by the sample makes multiple scattering important. In x-ray work this problem does not enter since the attenuation of the beam arises primarily from true absorption of the radiation. The effects of multiple scattering of neutrons in the sample can only be eliminated by making the sample very thin and then the peaks become too small to measure with the present available neutron intensities. We have, however, made approximate calculations of the amount of multiple scattering to be expected from our diamond sample and it agrees closely with the observed excess scattering.

On this basis then, it can be concluded that there is no measurable spin or isotope effect arising from the presence of the small amount of C^{13} in normal carbon and that the true diffuse scattering by the sample arises from the effects of lattice vibrations which can be corrected for by the usual Debye-Waller temperature factor which Weinstock[9] has shown to apply also to the case of neutron diffraction.

The Bragg scattering by diamond, when corrected for the lattice vibration effects, constitutes within the accuracy of our measurements the total scattering by carbon atoms. On this basis the measurements can be normalized to the true scattering cross section of carbon.

It seemed advisable in this connection to make an independent measurement of the total scattering cross section of the diamond powder by the transmission method. These measurements gave a value of 4.9 barns for the total scattering cross section by diamond powder at our particular neutron wave-length of 1.057A. This value is related to the scattering cross section for bound carbon atoms σ, according to the expression

$$\sigma_{\text{transmission}}=\frac{\lambda^2N}{8}\sum_{hkl} jd\sigma e^{-2W}R, \tag{19}$$

with the summation carried over all Bragg reflections and where

$$R_{hkl}=\sum_{\substack{\text{unit}\\\text{cell}}}\exp[2\pi i(hx+ky+lz)],$$

summed over all atomic positions (x, y, z) in the unit cell. N is the number of unit cells per cm^3, j the multiplicity value, and d the interplanar spacing value. Upon insertion of appropriate numerical values into Eq. (19) one obtains $\sigma=5.2$ barns. Since this corresponds to bound carbon atoms, the scattering cross section for free carbon centers is smaller by the reduced mass factor and thus $\sigma_f=4.4$ barns. This value is lower than the usually accepted carbon cross section of 4.8 barns, but it agrees well with the results of Rainwater and Havens.[17] The measured quantities $k^2f_0^2$ in Table I can now be normalized in terms of a bound carbon scattering cross section of 5.2 barns and thus k^2, which is a constant of our experimental arrangement, is evaluated. The average value of $k^2f_0^2$ for the measured peaks has been taken for use in determining k^2 in terms of the above cross section of carbon. Any later measurements can then be evaluated in terms of absolute cross sections by comparison with diamond which is taken as standard.

Graphite

Diffraction measurements with powdered graphite are shown in the upper part of Fig. 3. The scattering cross section for carbon (see Table I) has been determined from the (002) reflection alone, since the other peaks are less accurately measured. The intensity of this peak corresponds to a slightly higher carbon scattering cross section than the average value obtained from diamond, but the difference is within the experimental error.

Aluminum

The diffraction results for aluminum are shown in Fig. 4. The sample on which the measurements were made consisted of a ½-in. thick plate of pure rolled aluminum which was several times heated to near the melting point and then quenched. This process was found to remove preferred orientations in the sample and by using a solid sample more material can be exposed to the neutron beam and hence greater accuracy can be achieved.

The results of the measurements standardized against diamond are shown in Table II. The characteristic temperature for Al is sufficiently low (398°K) so that a very appreciable lattice vibration effect is observed over the angular range measured. One observes, however, that after application of the factor e^{2W} the Bragg scattering amplitudes f_0 are very nearly constant as is to be expected if the Weinstock treatment of the elastic scattering of neutrons is substantially correct.

The measured Bragg scattering cross section for aluminum agrees closely with the nuclear scattering cross section ($\sigma=1.48$ barns) and, since Al has only one isotope, it can be concluded

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[17] L. J. Rainwater and W. W. Havens, Jr., Report No. CP-1962, Manhattan District Plutonium Project.

that the scattering by aluminum has, if any, only a small dependence on spin.

Sodium Containing Crystals

In connection with the work on NaH and NaD reported in the following paper in this journal it is important to have an independent determination of the Bragg scattering amplitude of Na. Diffraction measurements on Na, NaBr, NaF, and NaCl, from which the Bragg scattering amplitude for sodium as well as for the other elements in the crystals can be determined, are reported here. Less extensive measurements have been made on these crystals than on those discussed above. The intensity patterns are shown in Fig. 5 and the results are listed in Table III. For NaBr, NaCl, and NaF the (111) and (200) reflections suffice to determine the amplitudes for the two elements in the crystal and for Na only one reflection (110) is required. With the compound crystals the usual powder sample was used. The measurements on Na were made with two samples; one consisted of the solid metal and the other consisted of small beads obtained by shaking the molten metal in xylene. The measurements from the two samples were in good agreement.

Fluorine

The measured Bragg scattering cross section for fluorine is found to be somewhat higher than the total bound nuclear scattering cross section, as taken from the literature. Actually, this cannot be the case and it must be ascribed to error in one or the other of the measurements. Since our measurements have been made on the first two peaks of a single fluorine compound, an uncertainty of 15 percent is not unreasonable. The results indicate, therefore, that there is only a small spin dependence, if any, for the scattering by fluorine.

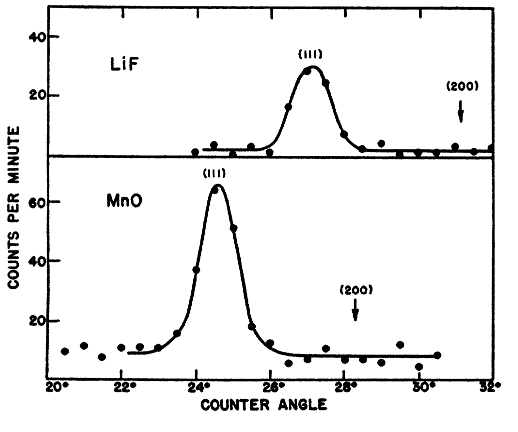

FIG. 6. Powder diffraction patterns for LiF and MnO. The absence of measurable (200) peaks shows that Li and F and also Mn and O scatter with opposite phase.

Bromine and Chlorine

Bromine and chlorine each have two isotopes and hence it is not expected that the Bragg scattering cross section should equal the nuclear scattering cross section.

From the values of σ and f with its proper sign and the isotopic abundances, the scattering amplitudes for the individual isotopes could be calculated by solving Eqs. (14) and (15) for f_1 and f_2. It is felt, however, that the measurements are of insufficient accuracy at present to warrant carrying out such a calculation.

Sodium

The scattering amplitude for Na is determined independently from each of the four crystals. The values of f_0 and $\sigma_B = 4\pi f_0^2$ for Na listed in Table III are seen to be in good agreement and they give an average value of $f_0 = 0.346 \times 10^{-12}$ cm and $\sigma_B = 1.51$ barns. The fact that σ_B is considerably smaller than the nuclear scattering cross section, $\sigma = 3.7$ barns, shows that the scattering by Na is spin dependent. The separate amplitudes for parallel and antiparallel spin can be calculated by solving Eq. (14) and Eq. (13), modified to the case of bound nuclei,* for $f_{i+\frac{1}{2}}$ and $f_{i-\frac{1}{2}}$, which gives

$$f_{i+\frac{1}{2}} = f_0 \pm \left(\frac{\sigma - \sigma_B}{4\pi} \times \frac{i}{i+1} \right)^{\frac{1}{2}},$$

and

$$f_{i-\frac{1}{2}} = f_0 \mp \left(\frac{\sigma - \sigma_B}{4\pi} \times \frac{i+1}{i} \right)^{\frac{1}{2}}. \qquad (20)$$

The data for sodium including the spin ($i = \frac{3}{2}$) when substituted into these equations give the

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* *Note:* σ_{coh} of Eq. (14) is the same as σ_B ($B \equiv$ Bragg) of Eq. (20). σ is used to designate the total scattering cross section by bound nuclei and σ_f is used for free scattering centers.

scattering amplitudes for the two spin states which are listed in Table IV. There is nothing in the experiments that permits one to make a choice between the two sets of values, but it might be argued that the set in which both amplitudes are positive is more apt to be the correct one.

The above value of 1.51 barns for the Bragg scattering cross section of sodium is not in agreement with the value of 3.5 barns reported by Fermi and Marshall.[1] Their measurements were made on the Bragg reflections from single crystals of NaCl with the chlorine cross section being arbitrarily adjusted to the total scattering cross section. Their intensity measurements were made by comparing peak heights rather than determining integrated intensities, and they have assumed that their crystals behave as if they were perfect (intensity proportional to first power of scattering amplitude). It would seem that these procedures might introduce considerable error in the calculated cross section values.

Phase of Nuclear Scattering

The sign of the phase shift of the total neutron wave relative to the incident wave determines the sign of the scattering amplitude. The relative sign of the scattering amplitudes for two elements can be readily determined by diffraction measurements on a crystal containing the two elements. With simple crystals such as NaCl, LiF, etc. a comparison of the intensities of the (111) and (200) peaks suffices for a comparison of the scattering phases since these two peaks correspond to a difference and a sum, respectively, of the scattering amplitude of the two elements. For more complicated crystals it may be necessary to measure more than two peaks to determine the relative scattering phases of two elements. Where an element consists of more than one isotope, the phase of scattering by the element will be determined by the predominant amplitude.

The powder patterns for the (111) and (200) peaks of LiF and MnO are shown in Fig. 6. The fact that the (200) peaks are smaller than the (111) peaks in LiF shows that Li and F have opposite scattering phases. The same is true of Mn and O, while for the Na compounds, shown in Fig. 5, the phases are all the same. Although one cannot from such comparisons alone determine the absolute sign of the phase shift, one can from other information readily identify the phase shift for Li and Mn as being positive.

Fermi and Marshall[1] have already published phase determinations for a number of elements. They use the convention of associating a positive scattering amplitude with a negative phase shift. Table V gives a list of elements for which the scattering phase has been measured either by us or by Fermi and Marshall; their terminology is used with respect to sign.

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