angular correlations and the structure of the correlation function. Secondly, the use of Eq. (8) is simpler than the use of the formulas and tables previously published, as the expansion in powers of the cosines of the angles is not the natural one, and makes formulas and tables much more complicated than necessary. For the use of Eq. (8) it is sufficient to tabulate separately the values of $c_{k\tau}(LL')$ for the different kinds of radiations, and the values of $W(jLjL';j_ik)$, which are independent of the kind of radiations, and to combine them according to necessity.[15] Tables of this kind are being prepared for publication.[16]

The author wishes to express his gratitude to Professor J. R. Oppenheimer for the generous hospitality in the stimulating atmosphere of the Institute for Advanced Study, and to Professor M. Fierz who suggested that he deal with this problem.

[15] Similar results have been obtained independently by S. P. Lloyd, Thesis, University of Illinois (1951); Phys. Rev. **80**, 118 (1950).

[16] J. M. Blatt and L. C. Biedenharn, Phys. Rev. **82**, 123 (1951).

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Neutron Scattering and Polarization by Ferromagnetic Materials

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Neutron diffraction studies are reported on a series of magnetized and unmagnetized ferromagnetic materials. The diffraction patterns for unmagnetized, polycrystalline samples of Fe and Co are found to possess both nuclear and magnetic components with the latter in agreement with the magnetic scattering theory with respect both to intensity of scattering and form factor angular variation. Studies on the magnetic structure of Fe_3O_4 are shown to strongly support Néel's proposed ferrimagnetic structure. Predictions of the theory regarding intensity effects upon sample magnetization are fully confirmed and the Schwinger-Halpern-Johnson formulation of the interaction function between the neutron's magnetic moment and the internal fields in a ferromagnet is substantiated. A pronounced variation of intensity around the Debye ring in the diffraction pattern for a magnetized sample is found. Neutron polarization effects in the Bragg scattered beams from magnetized crystals of Fe and Fe_3O_4 have been studied and it is shown that very highly polarized beams are obtained for certain reflections. This method of monochromatic beam polarization is found to compare very favorably with other methods with respect to polarization value, beam intensity, and ease of obtainment.

INTRODUCTION

THE theory of the scattering and polarization of neutrons by ferromagnetic substances has been given a very general treatment by Halpern and coworkers.[1] Up to the present time, experimentation in this field has centered upon studies of the single and double transmission effect[2] and more recently upon studies of critical reflection from magnetized mirrors.[3]

The present report deals with studies of the intensity distribution and the polarization of the scattered neutron radiation from both unmagnetized and magnetized ferromagnetic substances. These studies give information on the form factor dependence of magnetic scattering, on the basic nature of the neutron's magnetic interaction and on the magnetic structure existing in certain ferromagnetics, *viz.*, the spatial distribution of the various magnetic ions within the ferromagnetic lattice. A previous report[4] has discussed neutron scattering by paramagnetic and antiferromagnetic lattices and brief reports of some aspects of the present work concerning the neutron's magnetic interaction and the polarization phenomena have been given in the literature.[5]

The general expression for the differential scattering cross section of a magnetic ion has been given by Halpern and Johnson[1] (H-J) as

$$F^2=C^2+2CD\mathbf{q}\cdot\boldsymbol{\lambda}+D^2q^2 \qquad (1)$$

where C is the nuclear scattering amplitude, D the magnetic scattering amplitude, $\boldsymbol{\lambda}$ a unit vector describing the polarization state of the neutron being scattered,

$$\mathbf{q}=\mathbf{e}(\mathbf{e}\cdot\boldsymbol{\kappa})-\boldsymbol{\kappa} \quad \text{and} \quad q^2=1-(\mathbf{e}\cdot\boldsymbol{\kappa})^2 \qquad (2)$$

where $\mathbf{e}$ is the unit scattering vector and $\boldsymbol{\kappa}$ a unit vector parallel to the magnetic moment vector of the magnetic ion. The magnetic scattering amplitude D is given by

$$D=(e^2/mc^2)\gamma Sf=0.539\times10^{-12}Sf \text{ cm} \qquad (3)$$

[1] O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939). O. Halpern and T. Holstein, Phys. Rev. **59**, 960 (1941). Halpern, Hamermesh, and Johnson, Phys. Rev. **59**, 981 (1941).

[2] Hughes, Wallace, and Holtzman, Phys. Rev. **73**, 1277 (1948). Fleeman, Nicodemus, and Staub, Phys. Rev. **76**, 1774 (1949). Burgy, Hughes, Wallace, Heller, and Woolf, Phys. Rev. **80**, 953 (1950).

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

[4] Shull, Strauser, and Wollan, Phys. Rev. **83**, 333 (1951).

[5] Shull, Wollan, and Strauser, Phys. Rev. **81**, 483 (1951). C. G. Shull, Phys. Rev. **81**, 626 (1951).

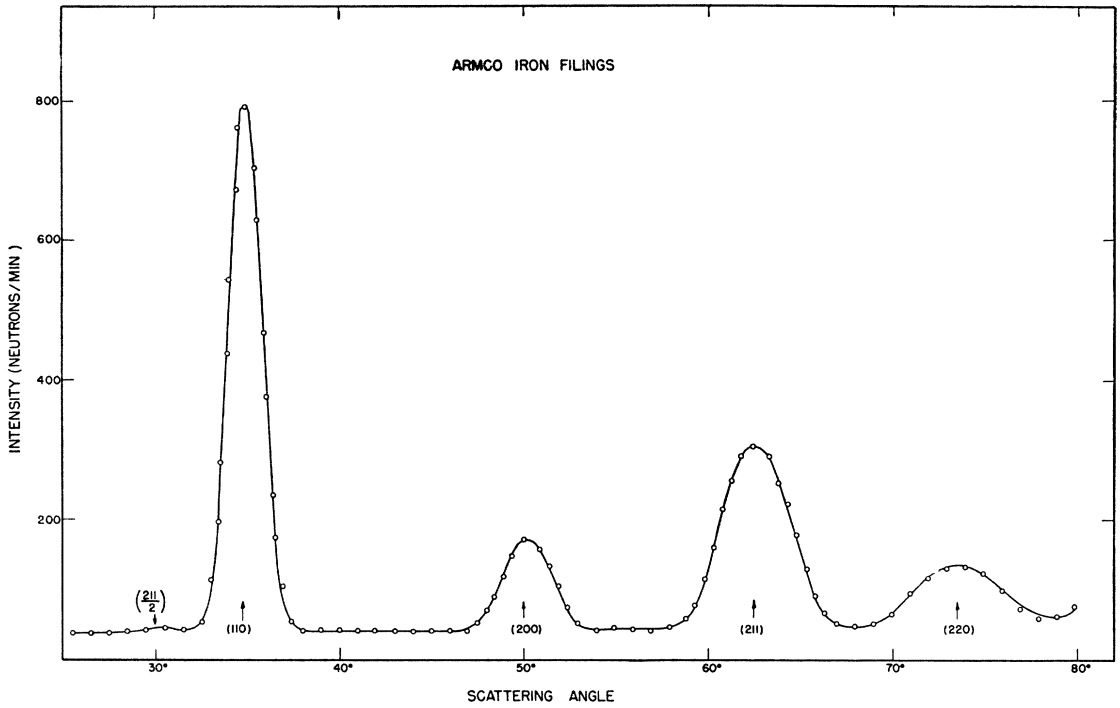

FIG. 1. Neutron diffraction pattern for polycrystalline Fe.

where γ is the neutron magnetic moment expressed in nuclear Bohr magnetons, S the spin quantum number of the magnetic atom, and f the magnetic form factor characteristic of the electrons responsible for the atomic magnetic moment.

For unpolarized incident neutron radiation, $\mathbf{q}\cdot\boldsymbol{\lambda}$ averages to zero and the differential cross section is the sum of the nuclear and magnetic contributions

$$F^2=C^2+D^2q^2. \tag{4}$$

The numerical value for q^2 will depend according to Eq. (2) upon the relative directions of the scattering vector and the magnetization vector. A further discussion of the form of this dependence will be given in a later section of this paper.

DIFFRACTION BY UNMAGNETIZED FERROMAGNETIC MATERIALS

We consider first the scattering by the ferromagnetic elements Fe and Co with no applied magnetic field. At temperatures below the Curie point, the magnetic moments of the atoms are in parallel alignment within a single domain but, without an applied field, the magnetic orientation from one domain to the next will, on the average, be random. Each domain constitutes a magnetic crystallite and since the magnetic moments are aligned within the domains, the magnetic scattering will appear in the Bragg peaks together with the nuclear scattering, the intensity of the two contributions being additive according to Eq. (4). The magnetic and nuclear contributions can be separately determined since the magnetic scattering will exhibit a form factor fall off with scattering angle due to the spatial distribution of the magnetic electrons within the atom while the nuclear scattering cross section is spherically symmetric. Following a discussion on the diffraction results for these ferromagnetic elements, data and their interpretation for a more complicated ferromagnetic substance, Fe_3O_4 (magnetite), will be given.

Iron

Figure 1 shows the powder diffraction pattern taken for a sample of polycrystalline Fe with neutrons of wavelength 1.204A. Rather high intensities are obtained for this material primarily because of the large coherent nuclear scattering amplitude for Fe. A number of samples were studied, with most emphasis on samples of filings taken from pure Armco iron blocks. The filings were given a heat treatment and hydrogen reduction to remove any oxide layer and some of the cold-working effects. The samples were contained in flat cells of thickness about $\frac{1}{2}$ in. between thin quartz windows. Corrections for second-order wavelength contribution were made to the patterns and aside from these low intensity second-order parasitic reflections, all of the observed reflections could be indexed on the basis of the usual body-centered cubic reflections for the normal cubic cell. The observed integrated intensities were placed on an absolute scale by comparison with a standard sample of nickel powder for which we have measured the coherent scattering cross section as 13.4 barns.

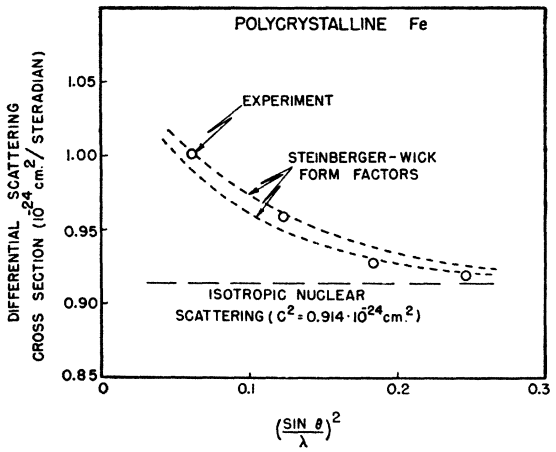

FIG. 2. Variation with angle of differential scattering cross section per Fe atom for the powder pattern reflections. The scattering cross section contains an angularly-dependent magnetic component and an isotropic nuclear component. The calculated magnetic scattering (with the Steinberger-Wick form factors) is shown.

The integrated intensities shown in the pattern of Fig. 1 were converted to differential scattering cross sections in the usual fashion[6] and these values for the four observed reflections are shown in Fig. 2. It is to be seen that this cross section shows a significant, although not pronounced, angular variation and this is because of the large isotropic nuclear scattering relative to the magnetic scattering. Using the magnetic moment per Fe atom of 2.22 Bohr magnetons as determined from saturation magnetization studies on iron, and hence a value of 1.11 for S_{eff} since the gyromagnetic ratio for Fe is very close to 2, one evaluates with Eq. (3) the expected magnetic scattering amplitude.

$$D(\text{Fe}) = 0.598 f \times 10^{-12} \text{ cm}. \qquad (5)$$

Steinberger and Wick[7] have given the results of a recent calculation of the magnetic form factor f for iron and on the basis of these values, the best fit between the data and Eqs. (4) and (5) is shown on the graph of Fig. 2. Also shown on the figure are extremes in the Steinberger-Wick calculation for different assumptions about the asymptotic behavior of the 3d-wave functions. The experimental data are not considered accurate enough to distinguish between one selection or another. Previous studies by Hughes, Wallace, and Holtzman[2] on the single transmission effect (change in transmission upon magnetization) with monochromatic neutrons have also shown general agreement with the magnetic scattering theory and the Steinberger-Wick form factor evaluation.

TABLE I. The coherent nuclear scattering cross sections of iron and its isotopes.

Isotope	Coherent scattering amplitude (10^{-12} cm)	Coherent scattering cross section (barns)
Fe^{54}	+0.42	2.2±0.1
Fe^{56}	+1.00	12.6±0.2
Fe^{57}	+0.23	0.64±0.05
Fe	+0.96	11.5±0.2

In the foregoing evaluation of the magnetic scattering cross section D^2 from the experimental data, q^2 has been taken as $\frac{2}{3}$ which is its average value when the magnetic moment within a domain has equal probability of all orientations relative to the crystal axes. It is generally agreed, however, that in iron the moments have a high probability of being aligned along the cube edges which are the easiest directions of magnetization in a single crystal. It can be shown, however, that even in this case the average value of q^2 will be $\frac{2}{3}$ for all (hkl) reflections with a polycrystalline and polydomain ferromagnetic sample. This fact makes it evident that the present data cannot be used to indicate the direction relative to the crystal axes of domain magnetization. This is in contrast to the conclusions[4] which have been drawn from antiferromagnetic reflections in MnO and FeO at low temperature wherein it was possible to establish the moment orientations even with polycrystalline data because incomplete multiplicities were obtained with these particular magnetic structures.

In addition to the studies of polycrystalline iron described above, a series of samples of isotopically enriched iron was also studied. Preliminary studies to determine the nuclear scattering phase of Fe^{54}, Fe^{56}, and Fe^{57} were carried out with the enriched iron in the form of Fe_2O_3, after which the metals in a finely divided state were studied in greater detail. For each of the isotopic iron samples, as for normal iron, the differential scattering cross section exhibited an angular variation. In the case of Fe^{56} the residual magnetic scattering after subtraction of the nuclear contributions was identical, within experimental error, with that obtained from normal iron, as indeed, is to be expected. A direct determination of the magnetic scattering from Fe^{54} and Fe^{57}, for which the nuclear scattering amplitudes are more favorable, did not prove feasible because of the very small amounts of sample at our disposal. In these two latter instances, the magnetic contribution was assumed known and equal to that for normal iron and Fe^{56}, and the nuclear scattering amplitude was obtained by subtraction. In Table I are shown the nuclear scattering properties of the isotopes of iron obtained in this way. The total scattering cross section of iron, using these isotopic cross sections, may be calculated and is found to be 11.6 barns. Comparison of this value with the coherent scattering cross section of 11.5 barns indicates that the isotopic diffuse scattering in normal iron is very small. This result supports the conclusions drawn by Hughes and Burgy[8] from their investigations of the single transmission effect in a single crystal of iron with neutron energies below that corresponding to

[6] C. G. Shull and E. O. Wollan, Phys. Rev. **81**, 527 (1951).
[7] J. Steinberger and G. C. Wick, Phys. Rev. **76**, 994 (1949).
[8] D. J. Hughes and M. T. Burgy, Phys. Rev. **80**, 481 (1950).

the first Bragg peak in which they find less than 0.25 barn of spin dependent or isotopic diffuse scattering.

COBALT

Diffraction patterns for a sample of face-centered-cubic cobalt in the form of metallic filings were obtained in the same fashion as in the above study on iron. The resulting scattering cross sections per magnetic atom are shown in Fig. 3 with the data resolved into its nuclear and magnetic contributions. Because of the much smaller nuclear scattering here than was found for iron, the magnetic scattering effects show up more strikingly.

Using the experimentally determined magnetic moment for Co of 1.74 Bohr magnetons[9] from magnetization studies on cubic cobalt, the magnetic scattering amplitude is evaluated as $0.468f\times10^{-12}$ cm. The value of the cross section corresponding to this amplitude at zero scattering angle (unit form factor) is shown on the ordinate axis of Fig. 4. The form factor describing the magnetic scattering is that suggested by the data and it is seen that this extrapolates satisfactorily to the calculated forward-direction cross section. It would be expected that this form factor would be closely similar to that calculated for iron by Steinberger and Wick and indeed this is substantiated in a quantitative comparison of the two. The slightly larger nuclear charge for Co would compress the 3d magnetic shell somewhat and the form factor would be slightly less angularly dependent than would be the case for iron. There is a suggestion of this in the Co experimental data.

As seen from the graph the coherent nuclear scattering cross section for Co amounts to only 1.0 barn and this is a consequence of the pronounced nuclear spin incoherent scattering for this nucleus. This has been discussed in an earlier report.[6] The magnetic scattering is comparable to or exceeds the nuclear scattering in the forward direction and this fact leads to interesting polarization phenomena in the neutron scattering by magnetized samples as will be discussed in a later section.

Fe_3O_4 (MAGNETITE)

This oxide possesses a spinel-type structure and is ferromagnetic on a macroscopic scale to about the same extent as nickel. There are both divalent and trivalent Fe ions present in the structure and the molecular formulation can be considered as containing one Fe^{++} ion and two Fe^{+++} ions. Verwey and Heilmann[10] consider the structure to be that of an inverted spinel in which the octahedral Fe sites are occupied at random by equal numbers of divalent and trivalent ions where as the tetrahedral sites are occupied only by trivalent ions. Further considerations by Néel[11] on the magnetic

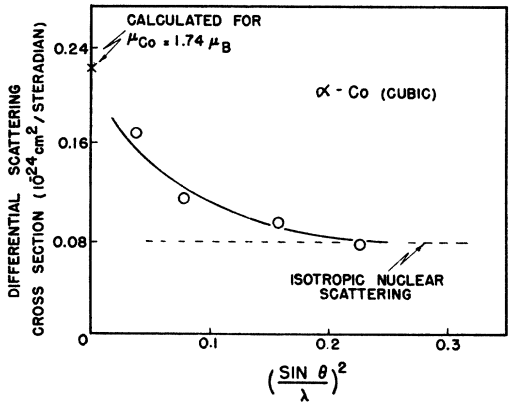

FIG. 3. Variation with angle of differential scattering cross section per Co atom for the Co powder pattern reflections.

properties suggest a ferrimagnetic structure for this material in which the magnetic moments of the tetrahedral ions are coupled antiferromagnetically to those of the octahedral ions. Because the latter are in the majority, Fe_3O_4 will be resultantly ferromagnetic and Néel has been able to explain quantitatively the observed magnetic moment per Fe atom obtained from magnetization data.

Since neutron scattering by such a magnetic lattice will be sensitive to the strength, position, and relative orientation of the magnetic ions, the neutron diffraction pattern should offer direct evidence as to the validity of these magnetic structure suppositions. Several powder samples have been examined, the principal quantitative measurements being made on a very pure sample kindly supplied by Professor A. von Hippel. A typical diffraction pattern taken at room temperature is shown in Fig. 3. As in the case of the simple ferromagnetic ele-

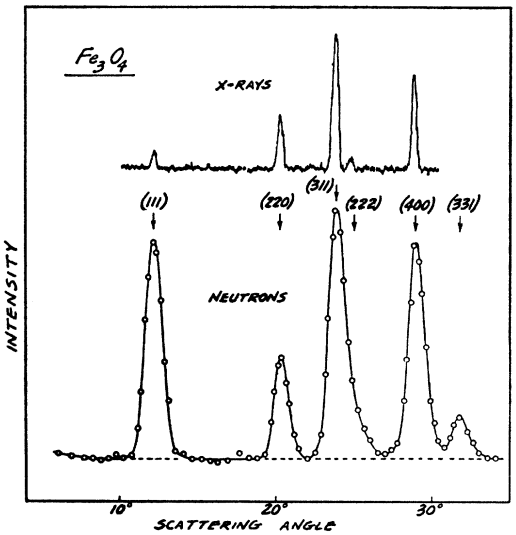

FIG. 4. Comparison between x-ray and neutron diffraction patterns for Fe_3O_4 (magnetite) at room temperature. Pronounced magnetic scattering contribution in the neutron pattern is found.

[9] A. J. P. Meyer and P. Taglang, Compt. rend. **231**, 612 (1950).
[10] E. J. W. Verwey and E. L. Heilmann, J. Chem. Phys. **15**, 174 (1947). Verwey, Haayman, and Romeijn, J. Chem. Phys. **15**, 181 (1947).
[11] L. Néel, Ann. phys. **3**, 137 (1948).

TABLE II. Comparison between observed intensities for Fe_3O_4 and those calculated for the Néel magnetic structure model.

Reflection	Crystal structure factor	Calculated intensities: Magnetic	Nuclear	Total	Observed intensity
(111)	$2(4f_1-4\sqrt{2}f_2)^2$	902	32	934	860
(220)	$(8f_1)^2$	125	218	343	360
(311)	$2(4f_1+4\sqrt{2}f_2)^2$	112	948	1060	1070
(222)	$(-16f_2+32f_0)^2$				
(400)	$(8f_1-16f_2-32f_0)^2$	116	649	765	780
(331)	$2(4f_1-4\sqrt{2}f_2)^2$	94	16	110	135
(422)	$(8f_1)^2$				
(333)	$2(4f_1+4\sqrt{2}f_2)^2$	20	670	690	700
(511)	$2(4f_1+4\sqrt{2}f_2)^2$				
(440)	$(8f_1+16f_2+32f_0)^2$	32	1692	1724	1730
(531)	$2(4f_1-4\sqrt{2}f_2)^2$				

All intensities (neutrons/min) are on an absolute scale.

ments already discussed, there will be no coherence between the nuclear and magnetic portions of the pattern. Since the nuclear scattering amplitudes for iron and oxygen are known accurately and the crystallographic structure has been well established from x-ray studies, the nuclear scattering contribution in the neutron pattern can be reliably accounted for. The remaining portion can be attributed to magnetic scattering and consequently can be compared to that predicted by any suggested magnetic structure.

The comparison between the observed and the calculated intensities for the Néel structural model is given in Table II along with the crystal structure factors which characterize the various reflections. In this tabulation, f_1, f_2, and f_0 represent the scattering amplitudes of a tetrahedral iron ion, an octahedral iron ion, and an oxygen ion respectively. For the nuclear scattering amplitudes, that for iron is taken as $0.956\cdot10^{-12}$ cm and for oxygen $0.575\cdot10^{-12}$ cm. On the Néel magnetic model,

$$(f_1)_{\text{mag}}=-D(\text{Fe}^{+++})$$

and

$$(f_2)_{\text{mag}}=+\tfrac{1}{2}D(Fe^{+++})+\tfrac{1}{2}D(\text{Fe}^{++}), \qquad (6)$$

with reversed signs for the two amplitudes because of the postulated antiferromagnetic orientation of tetrahedral and octahedral ions. The spin quantum numbers

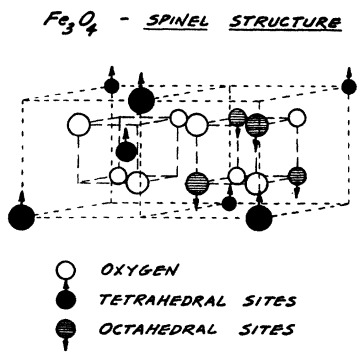

FIG. 5. Portion of the magnetic unit cell for Fe_3O_4. The octahedral and tetrahedral Fe ions are coupled antiferromagnetically.

for Fe^{+++} and Fe^{++} ions are, respectively, 5/2 and 2 and hence the appropriate magnetic scattering amplitudes are calculable from Eq. (3) as

$$\begin{aligned} D(\text{Fe}^{++})&=1.088f\times10^{-12}\text{ cm},\\ D(\text{Fe}^{+++})&=1.360f\times10^{-12}\text{ cm}. \end{aligned} \qquad (7)$$

Finally, the total intensity in an (hkl) reflection, say the (111) reflection, is taken as

$$I_{111}=I_{\text{nuclear}}+I_{\text{magnetic}}=k\{2[4f_{\text{Fe}}-4\sqrt{2}f_{\text{Fe}}]^2\}_{\text{nucl}}+\tfrac{2}{3}k\{2[4f_1-4\sqrt{2}f_2]^2\}_{\text{mag}} \qquad (8)$$

in which k is an instrumental parameter determinable from the powder diffraction formula. In the numerical evaluation, the magnetic form factor for Mn^{++} (reference 4) has been used for the Fe ions since the latter have not been established accurately at the present time. This should be a good approximation since the Mn ion and Fe ion would not be expected to differ much in their 3d-shell characteristics.

The intensities shown in Table II are all on an absolute scale and the agreement between calculated and observed total intensity values appears satisfactory. Magnetized sample studies to be discussed in the next section permit a direct resolution of the total intensity into the magnetic and nuclear components and these also support the validity of the Néel structural model. Other magnetic models have been investigated and it is found that the calculated intensity is rather sensitive to the selected model. For instance, an alternative structure would be for the Fe^{++} ions to exist wholly at tetrahedral sites and the Fe^{+++} only at octahedral sites (a normal spinel structure). The observed magnetization could be obtained for this model by having the Fe^{++} ions coupled ferromagnetically but with no orientation coupling between the Fe^{+++} ions themselves or between Fe^{+++} and Fe^{++}. Thus the trivalent ions would be in a paramagnetic state. Intensity calculations for this model are in violent disagreement with the data, with the (111) intensity for instance, being eight times lower than observed. Thus the Néel model of antiferromagnetic coupling between the tetrahedral ions and the octahedral ions in the inverted spinel structure appears fully substantiated. Figure 5 shows a portion of the magnetic unit cell of Fe_3O_4 with this antiferromagnetic arrangement of the ions.

Some diffraction data were also obtained for Fe_3O_4 at low temperatures (80°K) to see if any structural changes developed upon cooling. Fe_3O_4 exhibits interesting changes in magnetic properties and electrical conductivity at temperatures of the order 120–150°K and Verwey, Haayman, and Romeijn[10] have suggested that the pronounced increase in resistivity in lowering the temperature through 120°K is to be accounted for by an ordering of the Fe^{++} and Fe^{+++} ions at the octahedral sites. On this picture the relatively low resistivity at room temperature is caused by ionic migration (electron exchange) among the randomly

arranged ions at the octahedral sites. The neutron diffraction patterns at high and low temperatures were found to possess no significant difference (within about 1 percent) and a detailed examination of the crystal structure factors and the expected intensities for the Verwey model showed that this was not inconsistent with the model. The redistribution of the Fe^{++} and Fe^{+++} ions, which do not differ too much in magnetic scattering power (see Eq. (7)) does not appear to affect the powder intensities appreciably. If studies were to be made on single crystal, single domain samples where one does not have the superposition of various (*hkl*) reflections, then indeed the ordering effect should be noticeable but this has not been performed experimentally. The powder data do show, however, that the basic antiferromagnetic structure is maintained at low temperatures since any departure from ferromagnetic coupling between various octahedral ions, with consequent change of sign in the scattering amplitude, affects the calculated intensity pronouncedly.

SCATTERING BY MAGNETIZED SAMPLES

The magnetic contribution to the observed intensity of scattering in the diffraction pattern will depend according to Eq. (2) upon the relative orientation of the scattering vector and the ionic magnetic moment vector. Since the latter can be controlled in a ferromagnetic sample by application of a suitably high external magnetic field, there should be intensity changes when the field is altered either in strength or direction of application. By comparing the observed change with that calculated from the theory, it is possible to determine directly the magnetic component of the observed intensity and this would serve as an independent test on the magnetic scattering aside from the form-factor analysis discussed in the last section.

The diagrams shown in Fig. 6 illustrate the experimental arrangements of interest here. A monochromatic beam of neutrons is incident upon the polycrystalline ferromagnetic sample which is located within the pole gap of an electromagnet. The intensity of scattering in a portion of one of the Debye rings produced by the sample is studied (1) for the unmagnetized sample, (2) with the sample magnetized in a direction parallel to the scattering vector (arrangement (a) in the figure), and (3) with the sample magnetization perpendicular to the scattering vector [arrangement (*b*)]. According to Eqs. (2) and (4) the magnetic cross sections and hence intensities should have relative values for these three cases, $\frac{2}{3}$, 0, and 1, respectively.

For this purpose an electromagnet capable of producing a magnetic field of about 8000 oersteds in a 1-in. gap was mounted on the axis of the conventional powder diffraction spectrometer. The orientation of this field relative to the plane of scattering could be selected by suitable arrangement of the magnet. The state of magnetization of the sample was followed by search coil measurements of the magnetic field intensity at the surface of the sample. Normal sample size was about 1 in.$\times 1\frac{1}{2}$ in. in area and thickness $\frac{1}{4}$ in. or $\frac{3}{8}$ in. with the field application along the 1 in. direction.

The first measurements of this type were made with small blocks of pure Armco iron with studies of the (110) and (200) intensity as a function of magnetization strength and direction. It was found that the intensity of scattering was decreased relative to the unmagnetized sample value when the field was applied parallel to the scattering vector, as is expected from the theory. On the other hand, the intensity also decreased when the sample was magnetized perpendicular to the scattering vector, an effect which was not at first expected. The decrease in this case arises from the fact that in the scattering process for the case of field perpendicular to the scattering vector the neutrons are partially polarized and these will experience a different transmission cross section in their passage through the polycrystalline iron block. When the attenuation effects characteristic of the partially polarized beam produced upon scattering are properly taken into account, one calculates for the

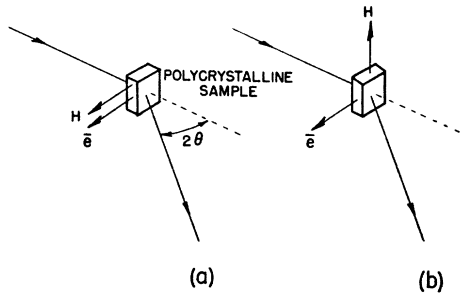

FIG. 6. Diagram of experimental arrangement used in the study of neutron scattering by magnetized, ferromagnetic substances. In (a) the applied magnetic field is in the plane of scattering and parallel to the scattering vector. In (b) the field is perpendicular to both.

(110) Fe reflection a decrease upon magnetization of 4.8 percent which agrees quite well with the experimentally observed value of 5.1 percent. Thus in spite of the larger cross section for scattering when $q^2=1$, the scattered beam is attenuated more in the sample and the expected intensity change is reversed.

The intensity effects which arise with iron upon magnetization are quite small as illustrated and this is because the magnetic scattering amplitudes are small in comparison to the unfavorably large nuclear scattering amplitude. More favorable cases are suggested for certain reflections of Co and Fe_3O_4 already discussed. The nuclear scattering amplitude for Co is quite small and should permit very large intensity effects upon magnetization but it is difficult to saturate magnetically. Magnetite on the other hand is rather easy to saturate magnetically and some of its reflections are very favorable for the present purpose. The (111) Fe_3O_4 reflection is almost completely magnetic in origin (see Table II) and this offers two advantages: (a) the inten-

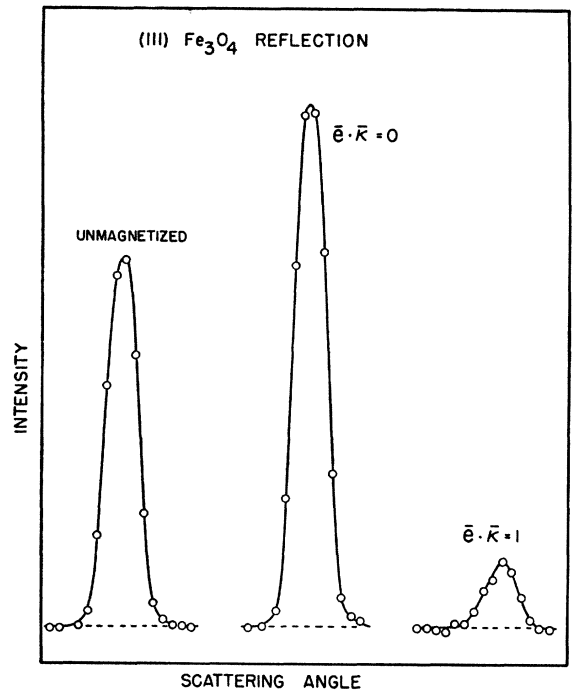

FIG. 7. Study of the (111) Fe_3O_4 reflected for different magnetization directions. The intensity is seen to depend strongly on the sample magnetization direction with respect to the scattering vector.

sity will be strongly affected by magnetization strength and direction and (b) there is very little polarization produced upon scattering, unlike the iron and cobalt cases, so that anomalous transmission effects will not be encountered. Because of these interesting and favorable properties, magnetite was studied in some detail.

Compressed powder samples of Fe_3O_4 were held between the pole pieces of the electromagnet and again field strength measurements were made with a search coil at the surface of the sample. Figure 7 shows typical traversals taken over the (111) Fe_3O_4 powder reflection with the sample block unmagnetized and when magnetized at high field strength in the two directions shown schematically in Fig. 6. It is seen that the scattered intensity varies pronouncedly with field direction. Figure 8 summarizes the data taken with varying field strength for the two field orientations and shows the saturation effects characteristic of increasing domain alignment with increasing field strength.

On Fig. 8 are shown the cross sections, $(C^2+\frac{2}{3}D^2)$, C^2, and (C^2+D^2) which correspond to the unmagnetized and two magnetized states. The nuclear cross section term C^2 is very much smaller than D^2 for this particular Fe_3O_4 reflection, in good agreement with the structure factor calculations outlined in an earlier section. Actually the nuclear scattering contribution to this reflection is even smaller than is shown in Fig. 8 because there is some second-order wavelength contamination from other reflections which accidentally contribute intensity to the position of measurement. In contrast to this distribution of intensity in the (111) reflection, Fig. 9 shows the intensity variation with field in the (220) reflection. For this reflection, the nuclear and magnetic contributions are about equal so that the intensity splitting is not as pronounced as for the (111) reflection. This close equality of nuclear and magnetic scattering in the (220) reflection allows for very interesting neutron polarization phenomena which will be discussed in a following section.

In all of the foregoing considerations, the agreement between experiment and theory was based upon the Schwinger[12] (H—J) formulation of the interaction vector **q**. The numerical value of this vector squared follows from Eq. (2) as

$$q^2=\sin^2\alpha, \tag{9}$$

where α is the angle between the scattering and magnetization vectors, **e** and **κ**. Prior to the Schwinger and (H—J) treatments, Bloch[13] had developed the interaction vector into the form

$$\mathbf{q}_0=\mathbf{e}(\mathbf{e}\cdot\boldsymbol{\kappa}) \tag{10}$$

with $q_0^2=\cos^2\alpha$. The difference between these two expressions for the angular dependence of magnetic scattering is related to the question as to whether the neutron in its passage through a ferromagnetic medium is affected by the magnetic field strength H or by the magnetic induction B. Ekstein has discussed this relationship recently[14] with particular reference to the interpretation of magnetized mirror experiments. The present experiments on the effect of magnetization direction upon the scattered intensity serves as a direct confirmation of the form of the magnetic interaction. After correcting the (111) Fe_3O_4 reflection for residual

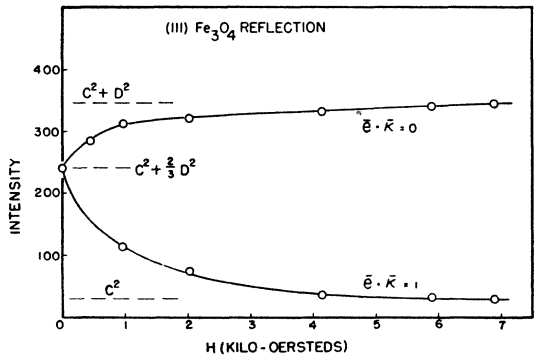

FIG. 8. Magnetic saturation for the (111) Fe_3O_4 reflection for different field directions. The variation with field strength illustrates the realignment of magnetization within the various domains into a direction parallel to the field. The nuclear scattering C^2 is seen to be very much smaller than the magnetic scattering for this reflection.

[12] J. S. Schwinger, Phys. Rev. **51**, 544 (1937).
[13] F. Bloch, Phys. Rev. **50**, 259 (1936); **51**, 994 (1937).
[14] H. Ekstein, Phys. Rev. **76**, 1328 (1949).

nuclear and second-order wavelength scattering, the remaining magnetic scattering cross section has been interpreted in terms of q^2 for four different experimentally-used values of the angle α: 0°, 23°, 54° (effective value for unmagnetized sample), and 90°. These values for q^2, normalized to the unmagnetized value of $\frac{2}{3}$, are shown in Fig. 10 along with the two theoretical curves. Within the experimental uncertainty of perhaps 2 percent the agreement with the Schwinger-Halpern-Johnson treatment is very satisfactory. Hughes and Burgy[3] in a recent study of critical reflection from magnetized mirrors have arrived at a similar conclusion although perhaps not within as close limits as in the present work.

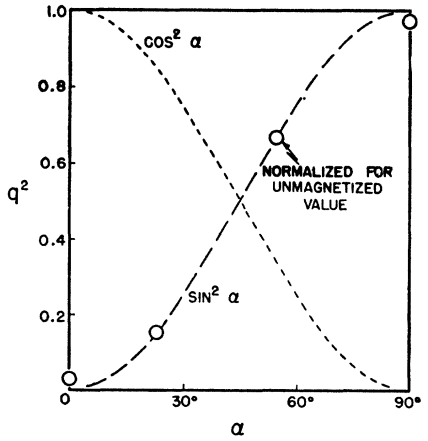

FIG. 10. Variation of q^2 with the angle α between the and scattering vectors.

POLARIZATION EFFECTS IN THE NEUTRONS SCATTERED BY MAGNETIZED SAMPLES

The commonly used method of producing a polarized neutron beam has been by transmission through highly magnetized samples of a ferromagnetic substance, usually pure iron, which possesses a different transmission cross section for the two characteristic spin states of the neutron. Supplementing this technique, Hughes and Burgy[3] have reported on the polarization effects associated with highly collimated neutron beams reflected from magnetized mirror surfaces. The present experiments are concerned with polarization of the neutrons diffracted by a crystal and are thus complimentary to the usual transmission experiments.

In the case of polarization by transmission, neutrons of one spin state are scattered out of the direct beam preferentially to those of the opposite spin state and a high degree of polarization is reached only asymptotically as larger and larger thicknesses are traversed. When one observes directly the neutrons diffracted by a small crystal it is theoretically possible to obtain complete polarization in a single scattering process. This follows from Eq. (1) in which, for the case of magnetization perpendicular to the scattering vector, $q^2=1$ and the differential cross section for unpolarized incident neutrons reduces to $(C\pm D)^2$. This is evident from the fact that an unpolarized beam can be resolved into two

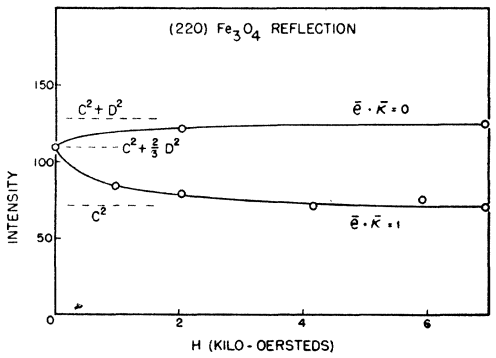

FIG. 9. Magnetic saturation for the (220) Fe_3O_4 reflection for different field directions. For this reflection the nuclear and magnetic scattering components are about equal.

equal and independent components of opposite spin state which can be arbitrarily taken parallel and antiparallel to the direction of the applied field. If the magnetic amplitude D and the nuclear amplitude C are equal for some reflection (hkl) of a crystal, the scattering will be zero for one spin state and the diffracted beam will be completely polarized. Lack of equality in the amplitudes will result in only partial polarization. As seen from Fig. 3 the first reflection from cobalt satisfies the condition for nearly complete polarization.

For those cases then where the nuclear and magnetic scattering amplitudes are known, it is possible to predict for a polycrystalline sample the degree of polarization for any given reflection and for any direction of observation relative to the direction of magnetization. The intensity of the diffracted beam from a powdered crystal is very low and hence it was considered of practical interest to see what degree of polarization could be achieved in the much more intense reflections from single crystals. The relation between the diffracted intensity and the atomic scattering amplitudes for a real single crystal depends strongly on the mosaic structure of the crystal and hence it is necessary to actually measure the polarization which can be obtained in a given case.

The polarization in the reflected radiation has been studied in two cases: the (110) reflection for Fe and the (220) reflection in Fe_3O_4. By calculation the maximum polarization to be expected in the former case is about 60 percent whereas it should be 100 percent in the latter case because of the close equivalence of the amplitudes as is evident in Fig. 9. The iron single crystal was prepared by the Bridgeman method from the melt by Mr. Frank Sherrill of this laboratory and contained 5 weight percent of silicon for phase stabilization. A thin slice of metal about $\frac{1}{2}$-in. round and $\frac{1}{8}$-in. thick was cut from the single crystal ingot with the Fe (110) plane in the surface of the slice. A natural crystal of magnetite, Fe_3O_4 of unknown purity, was used in the second examination again with a thin slice about $\frac{1}{16}$-in. thick

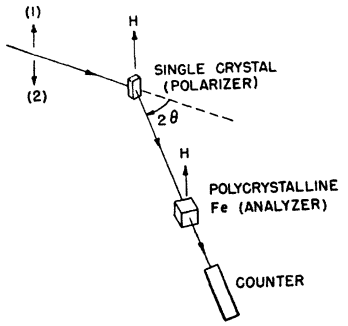

FIG. 11. Diagram of the experimental arrangement used in the study of neutron polarization produced by Bragg scattering from a magnetized crystal.

cut out along the (220) plane. Magnetization of the polarizing crystals was accomplished with an Alnico permanent magnet with H at the crystal surface about 4500 oersteds. The magnetization direction was within the plane of the crystal slices and along the [100] direction in both cases. Neutrons were reflected from internal planes within the crystal slice, the crystal being set in transmission orientation. A schematic diagram of the experimental arrangement is shown in Fig. 11.

The degree of polarization in the Bragg reflected neutron beam was studied by passing it through magnetized analyzing blocks of pure iron. Since the transmission cross section of the magnetized, analyzing block will depend upon the polarization, the latter can be evaluated if the properties of the analyzer are known. The analyzing block thickness was normally 1 in., although measurements were made with other thickness, and it was magnetized with an electromagnet to a field strength of about 8000 oersteds. The experimental procedure consisted of measuring the intensity of the polarized beam after passage through the analyzer when the latter was magnetized parallel to the polarizing fields and when unmagnetized. A magnetic field survey was conducted of the stray field between the polarizing and analyzing magnets and it was established that there was always a residual field of at least one hundred oersteds parallel to these two source fields. This situation sufficed then to insure that the neutron polarization was not altered in passing from polarizing region to analyzing region. Reversing the analyzing field with consequent depolarization effect on the beam was found to seriously effect the transmitted intensity as was to be expected.

Halpern and Holstein[1] have given general formulas describing the passage of a neutron beam through magnetized media, including the beam depolarization effects which arise when the medium is not magnetically saturated. Since the analyzing magnet in the present study was not of sufficient strength to saturate the analyzing block, it was necessary to take into consideration these depolarization effects. Their equations show that the depolarization can be allowed for in the polarization evaluation if measurements of the single transmission effect in the analyzing block are obtained with both the polarized beam and with an unpolarized beam. The latter was obtained by substituting a Cu single crystal in (111) reflecting orientation for the polarizing, ferromagnetic crystal.

In this method of polarization analysis, the nuclear and magnetic scattering amplitudes for Fe as determined in the earlier studies were used. Also in the theory application it was assumed that there existed no preferred orientation of crystallites in the polycrystalline analyzer and that the grain size was sufficiently small that the Debye-Scherrer diffraction rings were uniformly populated. This assumption, which has been implicitly accepted in all transmission studies to date, was found to be rather difficult to meet and is worthy of discussion. In studying the polarization produced in the (110) reflection from the magnetized Fe crystal, discordant results were obtained for various analyzing blocks cut from cold-rolled steel and Armco iron and further study showed that this was due to strong preferred orientation in the former and to large grain size (250μ) in the latter. As typical of these effects, there is shown in Fig. 12 the intensity distribution around the (110) Debye rings produced by two of the analyzing blocks with monochromatic incident neutron radiation. In all of the theoretical formulas developed for the polarization cross section it is assumed that this intensity distribution is uniform so that it is not surprising that the theory application would be poor for such an analyzer. Hughes and co-workers[4] have already pointed out that single transmission measurements are affected by orientation in the block although they do not appear

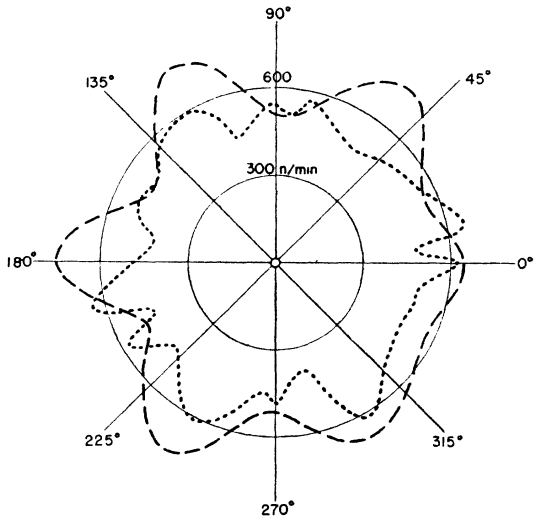

FIG. 12. Intensity variation around the (110) Fe Debye ring for two typical analyzing blocks of thickness about one centimeter. The dashed line is for a sample of cold-rolled steel (grain size about 25μ) and the dotted line is for an Armco block (grain size about 250μ).

to have taken it into account in their evaluation* of the polarization cross section for a thermal neutron distribution. It was found possible to obtain essentially complete randomness in grain orientation with a suitably small grain size (about 10μ) by solidification from the melt followed by repeated quenching of hot Armco blocks (900°C) in a dry ice-acetone mixture. This material was then cut into suitable analyzing blocks and these were used for studying the polarization in the Bragg beam from a polarizing crystal.

Table III summarizes measurements taken of the single transmission effect in a 1-in. thick analyzing block for the three different Bragg reflected beams. The unpolarized beam from a Cu crystal showed a +4.6 percent single transmission effect in the analyzing block and this served to establish the depolarization parameter in the analyzing block arising from incomplete magnetic saturation. Using the observed −3.0 percent effect with the polarized Fe beam, the polarization is calculated to be about 41 percent. This is somewhat smaller than the theoretical value for Fe (110) of 63 percent and may be caused by crystal extinction effects, silicon impurity in the crystal or lack of magnetic saturation in the polarizing crystal. All of these effects would tend to reduce the polarization from that predicted by the simple theory. The observed single transmission effect for the Fe_3O_4 (220) polarized beam is very much larger than for the other reflections, amounting to +24 percent and the evaluated polarization turns out to be 104 percent, which of course is impossibly high. It is felt that the measurements are accurate enough to say that the polarization is complete within about 5 percent which means that the relative neutron population in the two spin states is in ratio at least 40 to 1.

A very interesting further observation is to be noted in the data of Table III and this is the fact that the signs of the single transmission effect in the analyzing block for the Fe (110) and Fe_3O_4 (220) reflections are reversed. This indicates that the directions of polarization in the two polarized beams are opposite and hence that the atomic magnetic moments which are responsible for the polarization are directed oppositely with reference to the applied magnetic field on the two crystals. This is just the conclusion to be drawn from an analysis

* This may have contributed somewhat to the differences between the Stanford and Argonne Laboratory determinations of the polarization cross section for a thermal neutron beam (see reference 2). A large part of the difference appears to have arisen because of differences in the thermal neutron spectrum.

TABLE III. Comparison between experimental and theoretical neutron polarization in Fe (110) and Fe_3O_4 (220) reflections.

	Single transmission effect in analyzing block (1 in. thick)	Polarization calculated from data	Theoretical polarization for an ideally mosaic crystal
Unpolarized			
Cu (111)	+0.046±0.002	—	—
Polarized			
Fe (110)	−0.030±0.005	41%	63%
Fe_3O_4 (220)	+0.24±0.010	104%	100%

of the two magnetic structures. In the iron crystal all of the magnetic moments are aligned parallel to the applied field direction. In the magnetite crystal, on the other hand, the octahedral magnetic ions are aligned parallel to the field since they are more abundant than the antiferromagnetically coupled tetrahedral ions so that the latter are aligned antiparallel to the field. According to the crystal structure factors for magnetite given in Table II, only the tetrahedral ions contribute to the (220) reflection and hence the reversed polarization in this reflection is to be expected. The polarization observation for Fe_3O_4 can be considered a very direct proof of the antiferromagnetic nature of the magnetite structure. Interestingly there are other Fe_3O_4 reflections such as the (400) which should produce the conventional polarization sense as characterized by all of the metallic iron reflections.

It is possible to obtain approximately 10^5 monoenergetic neutrons per second in the (220) Fe_3O_4 reflection using neutron radiation from a pile with a central flux of 10^{12} neutrons/cm^2 sec. This compares very favorably in intensity with other methods of polarized beam production mentioned briefly at the beginning of this section. With regard to the three methods of polarization, *viz.*, (1) transmission through polycrystalline iron, (2) critical reflection from magnetized mirrors, and (3) Bragg scattering from ferromagnetic crystals, there appear to be unique characteristics for each and the selection of the optimum method should be decided by the requirements of the use to which the polarized beam is to be put.

ACKNOWLEDGMENTS

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