# Scattering of Neutrons by Spin Waves in Magnetite

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Measurements of energy distributions of 1.5 A neutrons diffusely scattered by a single crystal of magnetite in the region of the 111 reciprocal lattice point were carried out. Neutron groups were observed which satisfy momentum and energy conservation between the neutron and one wave-excitation quantum, and which are assigned a magnetic origin. The intensities of the neutron groups are consistent with spin wave theory within the limits of the analysis. The measurements are not sufficiently exact to enable the form of the frequency-wave number relation of the spin waves to be deduced, but assuming the quadratic relation of Kaplan a value for the A-B exchange integral of  $2 \times 10^{-3}$  ev is obtained.

#### INTRODUCTION

T is generally believed that the thermal disordering at low temperatures of the spin arrays of ferromagnetic or antiferromagnetic crystals can be described in terms of wave excitations called spin waves. The theory of the scattering of neutrons by spin waves<sup>1-3</sup> suggests that it should be possible to determine the relation between the energy of the spin waves and their wave number by studying energy distributions of the neutrons magnetically scattered by a single crystal. The neutron scattering is controlled by conservation of energy and momentum between the neutron and one spin wave quantum. If  $\hbar \mathbf{k}$  and  $\hbar \mathbf{k}'$  are the ingoing and outgoing neutron momenta, and E and E' are the ingoing and outgoing neutron energies, then

$$\mathbf{k} - \mathbf{k}' = 2\pi \tau \mp \mathbf{q} + \mathbf{w}, \tag{1a}$$

$$E - E' = \pm \hbar \omega, \tag{1b}$$

where **q** is the propagation vector of the spin wave,  $\hbar\omega$  is its energy,  $\tau$  is a vector of the reciprocal lattice and  $\tau + w/2\pi$  is a reciprocal space vector for which magnetic Bragg scattering occurs. For monatomic ferromagnetic substances w=0. These relations were established by Moorhouse<sup>2</sup> for simple ferromagnetic materials and by Elliott and Lowde<sup>3</sup> for several antiferromagnetic structures. It is likely that they are general.

From the energy distributions the energy and momenta of the observed neutrons can be deduced, and hence by Eqs. (1) the energy and propagation vector  $\mathbf{q}$ (or wave number  $q/2\pi$ ) of the spin wave quanta with which the neutrons interacted. By repeated observation the energy-wave number relation of the spin waves  $\omega(q)$  can be constructed.

To carry out the experiments a large single crystal is required of a substance in which the magnetic scattering is much larger than the nuclear scattering, and which has a Curie or Néel temperature high enough to make the neutron energy changes measurable with available resolution. It is also desirable that the nuclear capture and incoherent scattering be small. Magnetite (Fe<sub>3</sub>O<sub>4</sub>) fulfils these conditions and was thought to be the best possibility in spite of its comparatively complex ferrimagnetic structure.<sup>4</sup> Accordingly experiments have been performed in which energy distributions of neutrons scattered magnetically by a single crystal of magnetite at room temperature have been measured.

The results are consistent with Eqs. (1) indicating that wave-like spin excitations are indeed present in magnetite at room temperature. The intensities of the scattered neutrons are consistent with an intuitive extension to magnetite of the spin wave scattering theory of Elliott and Lowde.<sup>3</sup> The  $\omega(q)$  relation is discussed in terms of the mutually contradictory theories of spin waves in ferrites proposed by Kaplan<sup>5</sup> and by Vonsovski and Seidov.<sup>6</sup> For a simple ferromagnetic the energy of a spin wave is believed to be initially proportional to the square of the wave number<sup>7</sup> while for a simple antiferromagnetic the relation is believed to be linear.<sup>8</sup> For ferrites Kaplan<sup>5</sup> finds a quadratic relation, Vonsovski and Seidov<sup>6</sup> an essentially linear one. The uncertainties in our  $\omega(q)$  relation are such that we cannot distinguish between the two theories. However, fitting the experimental results to Kaplan's theory results in a reasonable value for the A-B exchange interaction integral of  $2 \times 10^{-3}$  ev, while the theory of Vonsovski and Seidov gives a value some five times greater. Thus these results favor Kaplan's quadratic relation.

<sup>&</sup>lt;sup>1</sup>G. Avakyants, J. Exptl. Theoret. Phys. 18, 444 (1948). <sup>2</sup>R. G. Moorhouse, Proc. Phys. Soc. (London) A64, 1097 (1951). <sup>3</sup>R. J. Elliott and R. D. Lowde, Proc. Roy. Soc. (London) A230, 73 (1955).

<sup>&</sup>lt;sup>4</sup> The unit cell contains 32 oxygen atoms, 8 Fe<sup>3+</sup> ions on "A" sites, 8 Fe<sup>3+</sup> and 8 Fe<sup>2+</sup> ions on "B" sites, arranged on interpenetrating F. C. C. lattices. The ions on the "A" sites are antiparallel to the ions on the "B" sites. See L. Néel, Ann. phys. 7, 710 (1948) and references 9 and 11 for details.

<sup>&</sup>lt;sup>6</sup> H. Kaplan, Phys. Rev. 86, 121 (1952). <sup>6</sup> S. V. Vonsovski and Y. M. Seidov, Izvest. Akad. Nauk S.S.R. 18, 319 (1954). Translation available through Columbia Technical Translations (5 Vermont Avenue, White Plains, New York). <sup>7</sup> F. Bloch, Z. Physik **61**, 206 (1930).

<sup>&</sup>lt;sup>8</sup> C. Herring and C. Kittel, Phys. Rev. 81, 869 (1951); J. M. Ziman, Proc. Phys. Soc. (London) A65, 540, 548 (1952); 66, 89 (1953); P. W. Anderson, Phys. Rev. 86, 694 (1952).

# EXPERIMENTAL

## Specimen and Its Contaminant Scattering

The specimen was cut from a natural single crystal of magnetite in the form of a flat plate about  $2\frac{1}{2}$  in.  $\times 1\frac{1}{2}$  in. $\times \frac{1}{8}$  in. thick containing about  $4.0 \times 10^{21}$  Fe<sub>3</sub>O<sub>4</sub> "molecules"/cm<sup>2</sup>. The edges were irregular due to cracking. Chemical analysis showed that the crystal contained 69.6% Fe by weight. Spectrographic analysis showed the presence of impurities as follows: 1.05% Ti, 0.25% Mn, 0.14% Al, 0.14% Si and 0.04% minor impurities. The formula of the crystal can be written as (Fe<sub>40.4</sub>Ti<sub>0.72</sub>Mn<sub>0.15</sub>Al<sub>0.15</sub>Si<sub>0.16</sub>Hole<sub>2.3</sub>)O<sub>58.5</sub> if all impurities and defects are assumed to be on the iron sites.

The crystal scatters neutrons in several ways which are of no interest in this experiment, except that the contaminant scattering must be separated from the magnetic inelastic scattering of interest. The contaminant scattering arises from two main sources, incoherent and thermal diffuse scattering and is considerably affected by multiple scattering.

In addition to 1.4 barns/Fe<sub>3</sub>O<sub>4</sub> "molecule" of nuclear incoherent scattering by the components, and 0.4 barn of magnetic incoherent scattering due to random distribution of  $Fe^{2+}$  and  $Fe^{3+}$  ions of the B sites in the crystal, there is incoherent scattering arising from random distribution of impurity atoms and holes in the crystal. Assuming the formula above, the nuclear part of this scattering amounts to 3.1 barns/Fe<sub>3</sub>O<sub>4</sub> "molecule." The magnetic part of this impurity disorder scattering cannot be computed without knowledge of the ionic states, but should not be greater than 3 barns per "molecule," the value if the impurities have zero moment. In the computation of magnetic scattering, the magnetic form factor of Hastings and Corliss<sup>9</sup> for the 111 reflection was used. The total incoherent scattering cross section is thus between 5 and 8 barns per "molecule." Since the angle of scattering was small. this incoherent scattering should be almost entirely elastic. The single thermal diffuse scattering was estimated<sup>10</sup> to be about 0.5 barn per "molecule" on the "independent vibrations" approximation.

The effect of multiple scattering was estimated from the following considerations. Multiple Bragg scattering can occur but rarely in a single crystal and then appears as sharp peaks in the angular and energy distribution, but after a neutron undergoes either an incoherent or thermal inelastic scattering, Bragg scattering can occur and, if extinction is neglected, almost the full scattering cross section is operative for rescattering. In this case the multiple scattering was estimated<sup>10</sup> to increase the incoherent scattering by about 20% of which half or more is expected to be elastic. By using the "independent vibrations approximation" for the inelastic scattering,<sup>10</sup> an estimate of (3 barns  $4\pi$  steradians) per Fe<sub>3</sub>O<sub>4</sub> "molecule" was obtained for the effective cross section for multiple thermal inelastic scattering in the forward direction. Since extinction was neglected, the figures of 20% and 3 barns are in the nature of upper limits. The total expected contaminant scattering is therefore roughly (10 barns  $4\pi$  steradians) per Fe<sub>3</sub>O<sub>4</sub> "molecule" of which approximately  $\frac{2}{3}$  is elastic, the remainder constituting a continuous widely-spreading inelastic spectrum.

#### **Experimental Method**

The experimental arrangement is shown in Fig. 1. Monoenergetic neutrons of mean wavelength  $1.52_3$  A were selected from the *NRX* reactor spectrum by Bragg reflection from the 111 plane of an aluminum single crystal not shown in Fig. 1. These monoenergetic neutrons impinged on the single crystal of magnetite which was oriented with its (011) direction vertical (normal to the plane of the spectrometers). Neutrons scattered at an angle of  $18._1^\circ$  were observed through a set of collimating slits  $C_1$ .

At an angle of scattering of 18.1°, Bragg reflection of 1.52<sub>3</sub> A neutrons by the 111 plane of magnetite occurs. By rotating the magnetite crystal the 111 Bragg peak is located. This peak consists<sup>11</sup> of about 97% magnetic scattering and only 3.5% nuclear scattering. By using  $C_2$  in line with  $C_1$ , the number of neutrons scattered was observed as a function of the angle  $\Psi$  by which the crystal deviated from the 111 Bragg scattering position. By comparison with the scattering by a standard specimen of vanadium metal, the differential cross section was obtained and is shown in Fig. 2. The dashed line shows the expected incoherent and multiple scattering as estimated in the preceding section. The strong scattering in the neighborhood of the Bragg position was observed previously by McReynolds and Riste<sup>12</sup> and is ascribed to magnetic inelastic scattering.

The energy distributions of these scattered neutrons were measured by means of a crystal spectrometer. The scattered neutrons pass through  $C_1$  and impinge on an aluminum single crystal. Another set of collimating slits  $C_2$  is set so that its angle with the 111 face of the aluminum crystal (analyzer) is always equal to the angle of incidence ( $\theta_A$ ) of the neutrons through  $C_1$ . The



FIG. 1. Schematic drawing of the apparatus. The  $01\overline{1}$  axis of the magnetite crystal is normal to the plane of the spectrometers.

<sup>&</sup>lt;sup>9</sup> J. M. Hastings and L. M. Corliss, Revs. Modern Phys. 25, 114 (1953). <sup>10</sup> See Brockhouse, Corliss, and Hastings, Phys. Rev. 98, 1721

<sup>&</sup>lt;sup>10</sup> See Brockhouse, Corliss, and Hastings, Phys. Rev. 98, 1721 (1955).

<sup>&</sup>lt;sup>11</sup> Shull, Wollan, and Koehler, Phys. Rev. 84, 912 (1951).

<sup>&</sup>lt;sup>12</sup> A. W. McReynolds and T. Riste, Phys. Rev. 95, 1161 (1954).

counter and collimator  $C_2$  rotate at an angular speed of one degree per hour, the analyzing crystal following at one half this angular speed. The counting rate was recorded as a function of the counter angle  $(2\theta_A)$  and the wavelength and energy of the scattered neutrons computed by the Bragg law. Background was determined by turning the analyzing crystal so that Bragg reflection into collimator  $C_2$  could not occur. Counts were recorded over 7.5 minute intervals with the analyzing crystal alternately in "signal" and "background" positions, corrected for pile power fluctuations, and each background point subtracted from the adjacent signal point. The resulting record with the magnetite crystal in various orientations is shown in Fig. 3. With this method of taking background the experiment is sensitive to very weak neutron groups, and it has been our experience that any nonstatistical fluctuations in the corrected counting rates are real.

The zero of the analyzing spectrometer angular scale and the wavelength of the incoming neutrons were determined by measuring the elastic scattering from vanadium metal13 in the first and second order of the analyzing crystal. The first order curve is used as a resolution function for the instrument.

The relative sensitivity function for the instrument has been obtained<sup>14</sup> by comparing the energy distribution of the scattering from the interior of a large clock of paraffin with a theoretical Maxwellian distribution. Over the range of interest in this experiment, the relative sensitivity function in a given order can be expressed as

$$\eta(\theta_A) = 0.150\theta_A - 1. \tag{2}$$

#### Results

Energy distributions were obtained with the magnetite crystal at orientations of  $\Psi = +20^{\circ}, +15^{\circ}, +10^{\circ},$ 



FIG. 2. Differential cross section per Fe<sub>3</sub>O<sub>4</sub> "molecule," at the angle of scattering for which Bragg scattering by the 111 plane of the crystal can occur, as a function of the deviation of the crystal from the Bragg position.



FIG. 3. Reduced counting rates with background subtracted, as a function of the angle of the counter of the analyzing spectrometer  $(2\theta_A)$  for various magnetite crystal orientations. Attention is drawn to the different ordinate scales. The dashed vertical line is at the incoming energy.

 $+5^{\circ}$ ,  $-5^{\circ}$ ,  $-10^{\circ}$ ,  $-15^{\circ}$ ,  $-25^{\circ}$ ,  $-35^{\circ}$ , and  $-40^{\circ}$  from the Bragg position. The results for a number of these orientations are shown in Fig. 3. The differences between signal and background counts, corrected for pile power fluctuations, are plotted as a function of the angle of the counter of the analyzing spectrometer  $(2\theta_A)$ . Auxiliary wavelength and energy scales are shown. Attention is called to the different ordinate scales for different orientations. For some of the curves, oddly shaped neutron groups were observed which were resolved into two groups with the characteristic resolution function shape.

At all the larger angles a neutron group of approximately constant intensity was observed at the incoming energy (elastic scattering). Comparison with the elastic scattering by the vanadium standard indicated a cross section 6.9 barns per Fe<sub>3</sub>O<sub>4</sub> "molecule" for this elastic scattering. This value is in agreement with the incoherent elastic scattering expected from the calculations of the specimen section.

The remainder of the pattern consists of inelastic neutron groups which are believed to be magnetic in origin. Their intensities decrease rapidly and the energy changes increase with increasing deviation from the Bragg angle. The energies and momenta of the observed neutron groups were analyzed in terms of Eqs. (1). In Fig. 4 the reciprocal lattice diagram corresponding

 <sup>&</sup>lt;sup>13</sup> B. N. Brockhouse, Can. J. Phys. 33, 889 (1955).
<sup>14</sup> B. N. Brockhouse (to be published).



FIG. 4. The reciprocal lattice diagram of the  $01\overline{1}$  plane. **k** and **k'** represent the incoming and outgoing neutron propagation vectors, the small circles represent the positions of the 111 reciprocal lattice points with the magnetite crystal in its various orientations. The crosses represent the termini of the **k'** vectors; the heavy lines represent the spin wave **q** vectors following Eq. (1). The component  $q_p$  of **q** in the direction of **k'** is indicated for one orientation. The resolution is indicated by the square insert.

to Eq. (1a) is shown. The incident neutron wave vector is **k**, the outgoing wave vectors **k'** corresponding to the various neutron groups are indicated by crosses. The 111 reciprocal lattice point with the crystal at the Bragg orientation is indicated by the circle labelled (111)<sub>0</sub>. The 111 reciprocal lattice point for other orientations ranges from the positions indicated by the circle labelled (111)<sub>20</sub> to that labelled (111)<sub>-40</sub>. The propagation vectors **q** are indicated by the heavy bars between the termini of **k'** (crosses) and the appropriate reciprocal lattice point.

The energies  $\Delta E = E - E'$  were computed for each neutron group and are plotted in Fig. 5 as a function of the magnitude of the wave number  $|\mathbf{q}|/2\pi$ . The errors shown are rough estimates of the uncertainties in q and  $\Delta E$  arising from the uncertainty in the positions of the neutron groups, and are intended to correspond to standard deviations. Errors in angles have not been considered but should be less than  $\frac{1}{4}$  degree and are therefore smaller than the errors considered above.

Resolution effects are very complicated. The resolution function in reciprocal space is indicated in Fig. 4, crudely estimated from the elements of the apparatus. The shaded area represents the  $\frac{1}{2}$ -intensity mark, the heavy line the limits of the resolution function. The observed neutron groups are, in addition, broadened by the energy resolution of the analyzing spectrometer. In the estimate no account was taken of an effect analogous to the parallel antiparallel crystal effect<sup>148</sup> well known in x-ray and neutron diffraction. This effect, which comes about because in Bragg scattering the angle of scattering and the wavelength are not independent variables, improves the resolution over that shown in Fig. 4. Some errors in q will be produced because different resolution elements have different scattering probabilities thus shifting the position of the mean. In particular it may be possible to satisfy the conservation laws only over part of the resolution function, i.e., only for some of the bundle of rays which represent the different incoming and outgoing neutron directions. The values of  $\Delta E$  thus can belong to considerably different q's than would be found simply by taking the q's corresponding to the nominal positions in the reciprocal space. This effect also reduces the intensity of the observed groups. It is thought to be important for the group at  $\Psi = 20^{\circ}$  as indicated by the dotted q in Fig. 4 and the point with arrows in Fig. 5. Additional measurements were made with the angle of scattering  $(\phi)$  increased by 1 degree. The results are

# DISCUSSION

not shown in detail but are included in Fig. 5.

The results described show that magnetic scattering of neutrons excites quantized wave excitations of some kind in magnetite at room temperature. The excitations seem to be of "acoustic" type for which  $\omega \rightarrow 0$  as  $q \rightarrow 0$ .

First it may be said that the excitations are probably not phonons<sup>15</sup> excited by way of the magnetic cross



WAVE NUMBER q/2 7

FIG. 5. The energy transfer  $\Delta E$  as a function of  $|\mathbf{q}|/2\pi$ , the "spin wave" wave number.

<sup>14a</sup> Note added in proof.—The specimen crystal was in the parallel position to the monochromating crystal (not shown in Fig. 1). The position of the analyzing crystal is not so important, and, as a matter of fact, the analyzing crystal was in the antiparallel position. Fig. 1 is in error in this respect.

<sup>15</sup> See R. Weinstock, Phys. Rev. **65**, 1 (1944); G. Placzek and L. Van Hove, Phys. Rev. **93**, 1207 (1954); B. N. Brockhouse and A. T. Stewart, Phys. Rev. **100**, 756 (1955); (to be published). section of the atoms-the so-called magneto-vibrational scattering.<sup>3</sup> The straight line of Fig. 5 corresponds to a velocity of  $\gtrsim 10^6$  cm/sec considerably greater than the mean velocity expected for sound waves in magnetite<sup>16</sup> from the specific heat.<sup>17</sup> Furthermore the observed intensities are over an order of magnitude larger than estimates of magnetovibrational scattering.<sup>18</sup> The excitations are thus apparently in the spin system itself.

The results are now examined to see to what extent the observed excitations correspond to the spin waves of the theory. The  $\omega(q)$  relation of Fig. 5 is compared with the spin wave theories of Kaplan and of Vonsovski and Seidov, to see if the values obtained for the exchange integrals are reasonable, and the intensities of the neutron groups of Fig. 3 are compared with calculations based on spin wave theory.

#### $\omega(q)$ Relation

Using the semiclassical theory of spin waves, Kaplan<sup>5</sup> and Kouvel<sup>19</sup> have found that the spectrum of spin waves in a ferrite consists of six branches<sup>20</sup> the lowest of which has an energy proportional to  $q^2$  and the higher of which have fairly large finite energies at q=0. Fitting Kaplan's equation (6) for the "acoustical" branch to the parabola of Fig. 5, using  $S_A = 2.5$  and  $S_B = -2.25$  for the spins of the ions on A and B sites, respectively, and assuming the exchange integrals between ions on Asites on the one hand  $(J_{AA})$  and B sites on the other  $(J_{BB})$  are zero, the exchange integral  $J_{AB}$  between ions on A and B sites is found to be  $2.0 \times 10^{-3}$  ev. Use of this value in a molecular field formula<sup>19</sup> for the Curie temperature leads to a value of 1050°K in good agreement with the experimental 850°K.

On the other hand, in the limit  $J_{AA}=J_{BB}=0$ , the equation of Vonsovski and Seidov<sup>6</sup> give  $J_{AB} = 10.5 \times 10^{-3}$ ev when fitted to the straight line of Fig. 5, leading to a Curie temperature larger by a factor of six then the experimental value.

In general Eqs. (1) can be satisfied only over a limited range of settings of the magnetite crystal. For the quadratic law of Fig. 5, Eqs. (1) can be satisfied only within the angular range indicated by the arrows in Fig. 2. The cross section is observed to fall off at about these positions, roughly as expected when resolution effects are included.

#### Intensities

If exchange interactions only are considered, the scattering cross sections for spin wave scattering in simple ferromagnetic structures and in three different simple antiferromagnetic structures<sup>3</sup> with random orientation of the magnetic domains can be put in the common form

$$\sigma_i(\mathbf{k} \rightarrow \mathbf{k}') = \frac{8\pi}{3} \left\{ \frac{N_\lambda}{N_\lambda + 1} \right\} \frac{k'}{k} \left( \frac{e^2 \gamma}{mc^2} \right)^2 f^2 \exp(-2W) \frac{g_i^2}{|J_i|} \quad (3)$$

per  $4\pi$  steradians per unit cell, where e and m are the charge and mass of the electron,  $\gamma$  is the magnetic moment of the neutron in nuclear magnetrons, f is the magnetic form factor of the ion, and  $\exp(-2W)$  is the Debye-Waller factor. The factor  $g^2$  is a structure factor which takes account of the reduction in the scattering by the relative phases of the magnetic atoms in the unit cell. For the cases treated by Elliott and Lowde, the ions scatter in phase and

$$g^2 = \frac{1}{nS} |\sum_{\text{unit cell}} S_j \exp(2\pi i \boldsymbol{\tau} \cdot \mathbf{R}_j)|^2 = nS,$$

where S is the spin of the ion and n is the number of ions per unit cell. For quantum absorption (neutron gain of energy),  $N_{\lambda} = \left[ \exp(\hbar \omega / k_B T) - 1 \right]^{-1}$  is used; for quantum emission (neutron loss of energy),  $N_{\lambda}+1$  is used. Equation (3) is the cross section for particular energy and momentum transfers which satisfy the conservation relations (1) for the *i*th spin wave branch, i.e., for one neutron group. The factor

$$J = 1 + \frac{\epsilon \hbar}{2E'} [\mathbf{k'} \cdot \operatorname{grad}\omega(\mathbf{q})],$$

where  $\epsilon$  has the value +1 for neutron energy loss and -1 for neutron energy gain, sums over the number of events satisfying Eq. (1). For a parabolic law,  $\hbar\omega$  $=\alpha \hbar^2 q^2/2M$  and  $J=1+\epsilon \alpha q_p/k'$ , where  $q_p$  is the component of q in the direction of the outgoing neutrons  $\mathbf{k}'$ , and M is the mass of the neutron. For a linear law,  $\hbar\omega = \beta q \hbar^2/2M$  and  $J = 1 + \epsilon \beta q_p/qk'$ .

For magnetite Eq. (3) has not been shown to hold but will be assumed to do so. Examination of the equations of motion of Kaplan<sup>5</sup> shows that, in the limits  $J_{AA}=J_{BB}=0$  and q=0, the spins on A and B sites precess in phase at equal angles to the z axis and with a phase angle of 180°. Thus, for q=0, the x and y components of spin have the same arrangement over the unit cell as the z component. Hence interference effects should be the same as for elastic scattering and,

<sup>&</sup>lt;sup>16</sup> The connection between the specific heat and the mean velocity of sound depends on the specific structure of the vibration spectrum. For example the assumption that there is one acoustic branch of each polarization for each Fe<sub>6</sub>O<sub>8</sub> unit leads to a value  $c \simeq 5 \times 10^5$  cm/sec.

<sup>&</sup>lt;sup>17</sup> R. W. Millar, J. Am. Chem. Soc. 51, 215 (1929), and reference 19.

<sup>&</sup>lt;sup>18</sup> Using the elastic magnetic structure factor in an equation

of I. Waller and P. O. Froman, Arkiv Fysik 4, 183 (1952). <sup>19</sup> J. S. Kouvel, Technical Report 210, Cruft Laboratory, Har-vard University, 1955 (unpublished); and Phys. Rev. **102**, 1489

<sup>(1956).</sup> <sup>20</sup> Kaplan looked only for solutions in which the phases of the spin waves were the same for all A sites on the one hand and for all B sites on the other except for the trivial  $\mathbf{q} \cdot \mathbf{R}$  phase factors. This restricted the number of branches found to two, whereas Kouvel found six, the number to be expected since the iron atoms in magnetite lie on six interpenetrating face-centered cubic lattices. The branch of interest here is the lowest energy "acoustical" branch for which the two authors agree.

TABLE I. Experimental and theoretical cross sections for the neutron groups in units of  $(10^{-24} \text{ cm}^2/4\pi \text{ steradians})$  per Fe atom.

						σ(theoretical) Eq. (3a)	
$\Psi$		$\Delta E$ ev	q  cm <sup>-1</sup>	$q_p cm^{-1}$	σ (expt.)	Quad- ratic law	Linear law
-35°	$^a_b$	0.0106 0.0134	$\substack{0.140\times10^8\\0.160\times10^8}$	$^{+0.08 imes10^8}_{-0.113 imes10^8}$	$0.4_{5}$ 0.5	1.2 0.8	2.2 2.2
-25°	$^a_b$	0.0068 0.0105	$\substack{0.145 imes10^8\ 0.119 imes10^8}$	$^{+0.131 imes10^8}_{-0.115 imes10^8}$	$\begin{array}{c} 0.8_5 \\ 0.8 \end{array}$	1.3 1.1	2.5 2.1
-15°	a b	$\begin{array}{c} 0.0031 \\ 0.0075 \end{array}$	$0.065 \times 10^{8}$ $0.132 \times 10^{8}$	$+0.060 \times 10^{8}$ $-0.130 \times 10^{8}$	$2.3_5$ $1.2_5$	$\begin{array}{c} 5.6 \\ 1.4 \end{array}$	5.0 3.1

guided by the work of Elliott and Lowde, we take<sup>20a</sup>

$$g^{2} = \frac{1}{n_{A}|S_{A}| + n_{B}|S_{B}|} \times |\sum_{jk} [S_{A} \exp(2\pi i \boldsymbol{\tau} \cdot \mathbf{R}_{A_{j}}) + S_{B} \exp(2\pi i \boldsymbol{\tau} \cdot \mathbf{R}_{B_{k}})]|^{2}, \quad (3a)$$

where the summation is over the unit cell and  $\eta_A = 8$ and  $\eta_B = 16$  are the number of A sites and B sites per unit cell.

The intensities of some of the neutron groups labelled a and b in Fig. 3 have been corrected by Eq. (2), converted to cross sections by comparison with the elastic scattering by a vanadium standard, and are given in Table I. The equation for the theoretical cross section is sensitive to the value of  $q_p$  so that it is possible to make comparison between experiment and Eq. (3) only for neutron groups for which  $q_p$  is comparable with or larger than the resolution function. The neutron groups of Table I are the only ones which at all satisfy this criterion as may be seen from Fig. 4.

Cross sections computed from Eqs. (3) by using the  $\alpha$  determined from Fig. 5 and values for  $q_p$  determined from Fig. 4 are shown in Table I. The agreement with the experimental cross sections is probably within the theoretical and experimental uncertainties. It has not been possible to obtain a basis for a structure factor from the work of Vonsovski and Seidov. Assumption

that Eq. (3a) holds leads to the intensities shown in Table I for the linear law.

## GENERAL DISCUSSION

As has been seen, the neutron results agree with expectations from the Curie temperature and on the whole favor the quadratic energy wave number relation of Kaplan rather than the linear relation of Vonsovski and Seidov. The specific heat of magnetite has been measured by Kouvel<sup>19</sup> and found to obey a  $T^{\frac{3}{2}}$  law as required by the quadratic relation but the value of  $J_{AB}$  deduced is only  $0.45 \times 10^{-3}$  ev, much lower than the value  $2 \times 10^{-3}$  ev obtained from the neutron measurements. The quadratic relation leads to a  $T^{\frac{3}{2}}$  law for the magnetization and the linear relation to a  $T^2$  law.<sup>6</sup> Pauthenet has found a  $T^2$  law to hold for the low-temperature magnetization of magnetite as well as of nickel and cobalt ferrite.<sup>21</sup>

For manganese ferrite, on the other hand, Pauthenet finds a  $T^{\frac{3}{2}}$  law to hold, and if the coefficient is fitted to an expression given by Kaplan<sup>5</sup> a value of  $0.35 \times 10^{-3}$  ev is found for  $J_{AB}$ , not in serious disagreement with the value deduced from the specific heat of magnetite when the lower Curie temperature of manganese ferrite (600°K vs 850°K for magnetite) is taken into account, but again much lower than estimated from the neutron scattering results and the Curie temperature.

In order to proceed further with the study of spin waves in ferrites, clarification of the theoretical situation is urgently needed. It is intended to continue the neutron scattering experiments to study the dependence of the scattering on the temperature and on the applied magnetic field. It is also hoped to return to the problem outlined herein with better resolution and accuracy.

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<sup>&</sup>lt;sup>20a</sup> Note added in proof.—Dr. R. J. Elliott informs me that he has shown that, within the limits of validity of spin wave theory, Eqs. (3) and (3a) describe correctly the neutron scattering by the acoustic spin wave modes.

<sup>&</sup>lt;sup>21</sup> R. Pauthenet, Ann. phys. 7, 710 (1952). The  $T^2$  fit for magnetite leans heavily on two measurements below the anomaly [see S. C. Abrahams and B. A. Calhoun, Acta Cryst. 8, 257 (1955)] at 119°K and is therefore of doubtful significance since all the other measurements are at room temperature and above. For the other materials, the temperatures at which the measurements were made were perhaps too high for comparison with theory to be significant.