

Field Dependence of Neutron Scattering by Spin Waves

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The energy distribution of neutrons scattered in the vicinity of the 111 reciprocal lattice point of magnetite have been measured with and without a magnetic field along the scattering vector. The intensity with a magnetic field was greater than with no field by a factor of 1.41 in approximate agreement with the theoretical value of 1.5, showing that the one-quantum spin wave scattering depends on the orientation of the spin system in the way predicted by theory. For spin waves of very low energy, competing effects of opposite sign apparently exist which render small the over-all effect of a magnetic field on the diffuse scattering.

INTRODUCTION

FROM the theory of scattering of neutrons by spin waves¹ it is expected that magnetically scattered neutrons should fall into groups in the energy distribution, satisfying the interference and energy conservation conditions

$$\mathbf{k}_0 - \mathbf{k}' = 2\pi\boldsymbol{\tau} - \mathbf{q}, \quad (1a)$$

$$|E_0 - E'| = \hbar\omega, \quad (1b)$$

where \mathbf{k}_0 and \mathbf{k}' , and E_0 and E' are the ingoing and outgoing neutron wave vectors and energies, respectively, $\boldsymbol{\tau}$ is a vector of the reciprocal lattice for the case which we will discuss, and $\hbar\omega$ and \mathbf{q} are the energy and wave vector of a single spin-wave quantum. For a ferrimagnetic substance it is expected²⁻⁵ that several spin-wave branches should exist, including an acoustic branch for which $\omega \propto q^2$ for small \mathbf{q} . Neutron groups have been observed⁶ in scattering around the 111 reciprocal lattice point in magnetite which satisfy Eqs. (1) with an energy wave vector relation and intensities⁷ consistent with spin wave theory.

According to the theory the intensity of the groups should depend on the applied magnetic field. The cross section for elastic scattering involves the time-independent z component of the spins and is proportional to $1 - (\mathbf{e} \cdot \boldsymbol{\kappa})^2$, where $\boldsymbol{\kappa}$ is a unit vector in the direction of magnetization and $\mathbf{e} = (\mathbf{k}_0 - \mathbf{k}') / |\mathbf{k}_0 - \mathbf{k}'|$ is the unit scattering vector.⁸ Spin wave one-quantum inelastic scattering, on the other hand, involves the x and y components of spin and has intensity proportional to

$1 + (\mathbf{e} \cdot \boldsymbol{\kappa})^2$. If, by application of a field, the direction of magnetization $\boldsymbol{\kappa}$ is made parallel to the scattering vector \mathbf{e} , the quantity $1 - (\mathbf{e} \cdot \boldsymbol{\kappa})^2$ is reduced from its average value $\frac{2}{3}$ to zero, and $1 + (\mathbf{e} \cdot \boldsymbol{\kappa})^2$ is increased from its average value $\frac{4}{3}$ to 2.

However, Riste and Janik⁹ reported that the scattered intensity in the region of the 111 reciprocal lattice point in magnetite, believed to be largely one-quantum magnetic inelastic scattering, did not change upon application along the scattering vector of a field sufficient to saturate the crystal. This lack of response to an applied field was very disturbing because the spin orientation behavior described above is firmly rooted in spin wave theory. Accordingly we repeated some of our previous experiments with provision for an applied field, as described herein, with results in agreement with spin wave theory. Since carrying out these experiments we have received word of additional results by the JENER group.¹⁰ Under experimental conditions differing from those used previously, they now observe positive effects on application of a field, and attribute their previous null result to cancellation by competing effects of opposite sign.

EXPERIMENT

Energy distributions of 1.52 Å (0.035 eV) neutrons scattered by a natural single crystal of magnetite were measured in the same way as before using crystal spectrometers to provide the initial monoenergetic neutrons and to analyze the scattered neutrons. The specimen crystal was an approximate cylinder $\frac{5}{8}$ in. in diam by 3 in. long, oriented with its $[01\bar{1}]$ direction along the cylindrical axis, which was perpendicular to the plane of \mathbf{k}_0 and \mathbf{k}' . With the angle of scattering $\phi = 18^\circ$ and the crystal properly oriented, Bragg scattering by the $\{111\}$ planes occurred. A magnetic field of nominal value $H = 3500$ oersteds was applied perpendicular to the direction of \mathbf{k}' and in the $(01\bar{1})$ plane. With the crystal oriented at 10.2° from the Bragg reflection position¹¹ ($\Psi = -10.2^\circ$), H lay along the scattering vector \mathbf{e} and in the easy $[111]$ direction of

¹ See R. J. Elliott and R. D. Lowde, Proc. Roy. Soc. (London) **A230**, 73 (1955).

² H. Kaplan, Phys. Rev. **86**, 121 (1952).

³ J. S. Kouvel, technical report, 210 Cruft Laboratory, Harvard University, 1955 (unpublished).

⁴ T. A. Kaplan, Phys. Rev. **109**, 782 (1958).

⁵ Kondorski, Pakhamov, and Shiklosh, Proc. Acad. Sci. U.S.S.R. **109**, 931 (1956), Soviet Phys. Doklady **1**, 501 (1956).

⁶ B. N. Brockhouse, Phys. Rev. **106**, 859 (1957).

⁷ The structure factors given in reference 6 for the antiferromagnetic and ferrimagnetic unit cells are in error, the error in the latter case amounting to about a factor of 3. This does not destroy the approximate agreement between theoretical and experimental cross sections (reference 6, Table I) because a compensating numerical error of about the same amount was made in calculating the theoretical cross sections. The author is grateful to Dr. T. A. Kaplan for pointing out these mistakes.

⁸ O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939).

⁹ T. Riste and J. Janik, JENER Report No. 53, November, 1957 (unpublished).

¹⁰ Riste, Blinowski, and Janik (private communication).

¹¹ See reference 6, Fig. 3.

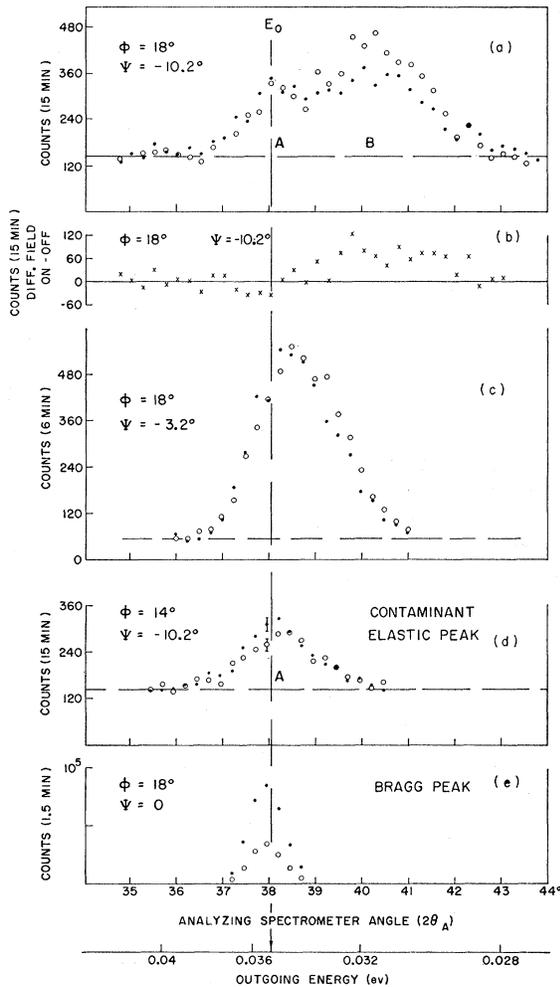


FIG. 1. Energy distributions of 0.035-eV (1.52 Å) neutrons scattered by a single crystal of magnetite as a function of the analyzing spectrometer angle for various angles of scattering (ϕ) and for various orientations (Ψ) of the specimen crystal. Distributions without field are shown by closed circles, with field by open circles, and field on-off differences by crosses.

magnetization of the crystal. The crystal is believed to have been saturated upon application of the field.

Energy distributions were taken [using a (111) plane of an aluminum crystal in the analyzing spectrometer as previously described], with and without an applied field and are shown in Fig. 1(a). Before making the measurements without applied field the crystal was accommodated by repeated application of the field in another direction [111]. This procedure was adopted to eliminate any possibility that a 180° domain system had been set up in the [111] direction by previous applications of the field. [Note that the neutron intensity is sensitive only to the directions of the spin vectors and not to their sense.] The background shown by the dashed line was obtained by turning the analyzing crystal out of the Bragg position, and then

adding a small correction for contaminant inelastic scattering. The intensity of the spin wave neutron group B is seen to have increased by a factor of 1.40 ± 0.05 upon application of the field. The experiment was repeated, but with the field applied and removed at each position of the analyzing spectrometer. The differences between the counts with and without field are plotted in Fig. 1(b). The increase observed with field corresponds to a factor 1.42 ± 0.05 in satisfactory agreement with the factor of 1.5 expected on spin wave theory under perfect conditions.

The published experiment of Riste and Janik was made with a mix-setting of 3° at the same wavelength and angle of scattering as these experiments. Accordingly, with the specimen at 3.2° from the Bragg angle ($\Psi = -3.2^\circ$), energy distributions were taken with field applied and field off, point-by-point. The results are shown in Fig. 1(c). The integrated intensity is greater with field on than with field off by a factor of only 1.04 ± 0.02 , in substantial agreement with the null result of Riste and Janik. It will be observed, however, that an apparent shift in the energy of the peak occurred. This shift and the nearly null over-all effect can be accounted for as a superposition of effects of opposing sign. Experiments under higher resolution could distinguish between the various possibilities: contaminant Bragg scattering, multiple quantum scattering, magnetovibrational scattering, and change of spin wave energy with applied field. Experiments under high resolution to obtain the dispersion $\omega(\mathbf{q})$ relation accurately are planned.

The response of the Bragg scattering by the (111) plane, and of the contaminant elastic incoherent scattering, to the applied field was also measured. Ideally the magnetic part of this scattering should disappear if the crystal were completely saturated by a field along the scattering vector, as has been repeatedly observed by other workers. The Bragg scattering was in fact reduced by 60% upon application of the field as seen in Fig. 1(e). The calculated reduction, including the effect of the nuclear scattering and of deviation of the field direction from the scattering vector, is 95%. The difference is thought to arise from the effect of extinction in the large and comparatively perfect crystal used. The elastic incoherent contaminant peak A was isolated from the inelastic scattering by changing the angle of scattering to 14° . Energy distributions taken with and without applied field, point-by-point, are shown in Fig. 1(d). The elastic peak was reduced by a factor 0.85 ± 0.07 on application of the field, thus probably confirming that magnetic incoherent scattering contributes to the contaminant elastic scattering as previously discussed.⁶

The results described herein complete the identification of the excitations previously observed as spin waves of the usual description.