

## Dispersion Relation for Spin Waves in a fcc Cobalt Alloy

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The technique of neutron spectrometry has been used to measure the dispersion curve of the spin waves in a metal for the first time. The momentum distributions of inelastically scattered neutrons from a single crystal of fcc cobalt containing 8% of iron have been observed under constant energy transfer conditions. The observed neutron groups (which satisfied momentum and energy conservation between the neutrons and the spin-wave quanta) enabled the dispersion relation to be established. The response of the intensity of the neutron groups to an applied magnetic field was used to identify those of spin-wave origin. The dispersion relation is in agreement with the form predicted by the

Bloch-Heisenberg spin-wave theory. Over the range of measurements, which was limited by the available spectrum of neutron energies, approximate isotropy was observed to hold. The value of the product of the exchange integral and the atomic spin is found to be  $JS = (1.47 \pm 0.15) \times 10^{-2}$  ev. Assuming  $S = 0.92$  the value for  $J$  is in poor agreement with a value obtained from the spin-wave interpretation of low-temperature magnetization data. Study of the widths of the neutron groups leads to the conclusion that the mean lifetimes of some of the spin waves are greater than  $3 \times 10^{-13}$  sec.

### INTRODUCTION

THE dispersion curve of the spin waves in a ferromagnetic metal has been measured using neutron spectrometry. The spin waves were observed in a single crystal of an alloy of cobalt containing 8% of iron, by the analysis of inelastic neutron scattering from the crystal. The measured energy-momentum relation of the excitations is of the form predicted by the Bloch-Heisenberg spin-wave theory.<sup>1</sup> The value of the exchange integral deduced from the dispersion curve, however, is in poor agreement with the value deduced from low-temperature magnetization measurements.

The inelastic scattering of neutrons resulting from the creation or annihilation of a single spin-wave quantum (magnon) is subject to the conservation of energy and momentum. If the initial and final neutron propagation vectors are  $\mathbf{k}_0$  and  $\mathbf{k}'$ , respectively, and the associated energies are  $E_0$  and  $E'$ , then for a ferromagnetic crystal<sup>2</sup> the conservation relations

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

$$\Delta E = E_0 - E' = \pm \hbar\omega \quad (1b)$$

hold, where  $\mathbf{q}$  is the propagation vector of the spin wave and  $\hbar\omega$  is its energy.  $\boldsymbol{\tau}$  is a reciprocal lattice vector.

If the neutrons which suffer an energy loss  $\Delta E$  are observed as the crystal orientation  $\psi$  and scattering angle  $\phi$  are changed, a neutron group will be seen as the spin-wave vector satisfies Eqs. (1). The center of this group will give the values of  $\hbar\omega$  and  $\mathbf{q}$  of the spin wave which has taken part in the scattering process. A series of such observations employing a range of values of  $\Delta E$  establishes the energy-wave number relation  $\omega(\mathbf{q})$  of the spin waves.

Previously this technique has been applied only to magnetite<sup>3</sup> though neutron scattering involving inter-

action with spin waves has been studied in magnetite<sup>4</sup> and hematite.<sup>5</sup> The only metal in which spin waves had been observed was iron<sup>6</sup> for which the spin-wave interpretations of the neutron scattering data and low-temperature magnetization data were consistent. However, the work on iron involved only spin waves of very low energy, and no dispersion curve was obtained.

### THE EXPERIMENT

The face-centered cubic cobalt alloy, which was chosen for its phase stability at room temperature, has a Curie temperature  $T_c = 1300^\circ\text{K}$  and a Bohr magneton number ( $p_B$ ) of 1.84.<sup>7</sup> These properties together with the small coherent nuclear scattering cross section make the alloy suitable for study by this technique. The crystal was cut in the form of a plate (4 in.  $\times$  1 $\frac{1}{4}$  in.  $\times$   $\frac{1}{8}$  in.) and the long dimension coincided with the  $[0,1,1]$  direction so that the face was the (2,0,0) plane. It was mounted with the (0,1,1) plane horizontal. Monoenergetic neutrons of energy  $E_0$  were selected from the reactor spectrum by Bragg reflection from the (1,1,1) or (2,0,0) planes of an aluminum crystal and after collimation were scattered by the specimen. Those neutrons undergoing an energy loss  $\Delta E$  were counted as the angles  $\phi$  and  $\psi$  were varied in a predetermined way with  $E_0$  and  $E'$ , i.e.,  $|\mathbf{k}_0|$  and  $|\mathbf{k}'|$  held constant (Fig. 1). The outgoing neutrons of energy  $E'$  were detected by an analyzing spectrometer using the (1,1,1) plane of aluminum. In all cases magnon creation was employed, i.e.,  $E_0 > E'$ . The above procedure was adopted in order to obtain easily recognizable neutron groups with the extremely steep dispersion curve involved. The apparatus and methods are described in detail elsewhere.<sup>8</sup>

<sup>4</sup> T. Riste, K. Blinowski, and J. Janik, *J. Phys. Chem. Solids* **9**, 153 (1959).

<sup>5</sup> J. A. Goedkoop and T. Riste, *Nature* **185**, 450 (1960).

<sup>6</sup> R. D. Lowde, *Phys. Rev. Letters* **4**, 452 (1960); *Proc. Roy. Soc. (London)* **A235**, 305 (1956).

<sup>7</sup> P. Weiss and M. Forrer, *Ann. phys.* **12**, 359 (1929).

<sup>8</sup> B. N. Brockhouse, in *Proceedings of the Symposium on Inelastic Scattering in Solids and Liquids*, Vienna, October, 1960 (unpublished).

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<sup>1</sup> F. Bloch, *Z. Physik* **61**, 206 (1930).

<sup>2</sup> R. G. Moorhouse, *Proc. Phys. Soc. (London)* **A64**, 1097 (1951); R. J. Elliott and R. D. Lowde, *Proc. Roy. Soc. (London)* **A230**, 46 (1955).

<sup>3</sup> B. N. Brockhouse, *Phys. Rev.* **106**, 859 (1957); **111**, 1273 (1958).

Using the reciprocal lattice representation of Eq. (1a) the spin-wave vector  $\mathbf{q}$  is represented by the line drawn from the terminal point of  $-\mathbf{k}'$  to the nearest reciprocal lattice point. The angles  $\phi$  and  $\psi$  were chosen such that the terminal point of  $-\mathbf{k}'$  lay on a line which represented a chosen direction through the reciprocal lattice point, i.e.,  $\mathbf{q}$  was along a chosen direction. Distributions were taken as the magnitude of  $\mathbf{q}$  was changed by varying  $\phi$  and  $\psi$ . Linear variations were found to be adequate for this purpose.<sup>8</sup> The momentum distributions so observed revealed neutron groups and the origin of each was determined by the response of the neutron intensity to the application of a magnetic field.<sup>2-4</sup> A field of 5 koe was applied in a direction perpendicular to the scattering vector (i.e., along the  $[0,1,1]$  direction) and would be expected to result in a decrease in spin-wave scattering of 25% and an increase of 30% for phonon scattering which partially results from the creation of a phonon through the magnetic scattering cross section (magnetovibrational scattering). The distribution shown in Fig. 1(b) is the number of neutrons losing 0.00921 ev as the terminal point of  $-\mathbf{k}'$  was moved, as shown, in the  $[0,1,1]$  direction of the reduced zone through the  $(2,0,0)$  reciprocal lattice point by changing the angle  $\psi$ . The two spin-wave peaks showing the expected field dependence, are symmetrical within the errors, about the reciprocal lattice point. The position of the magnetovibrational peak leads to a value of the velocity of sound which is in good agree-

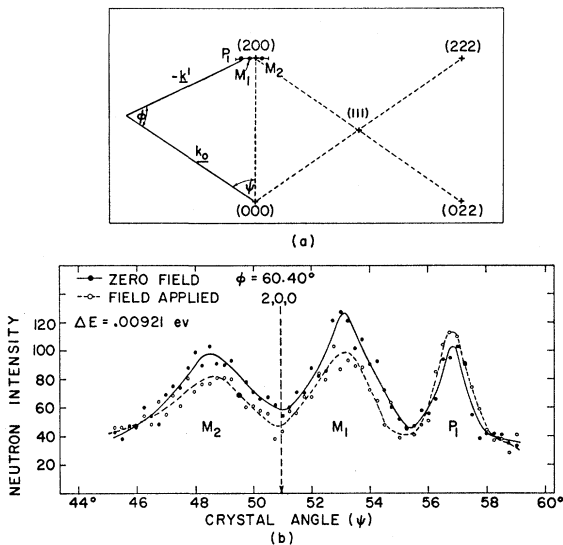


FIG. 1. (a) The  $(0,1,1)$  plane of the reciprocal lattice of fcc cobalt (+8% Fe) showing a typical arrangement of the vectors  $-\mathbf{k}'$  and  $\mathbf{k}_0$  when the crystal orientation  $\psi$  was changed so that the magnitude of the spin-wave vector varied in the  $[0,1,1]$  direction through the  $(200)$  reciprocal lattice point. (b) The distribution of neutrons losing energy  $\Delta E = 9.21 \times 10^{-3}$  ev as  $\psi$  was varied as in (a), for the case of zero applied field and of a field applied perpendicularly to the scattering vector.  $M_1$  and  $M_2$  represent the creation of a magnon and  $P_1$  represents the creation of a phonon.

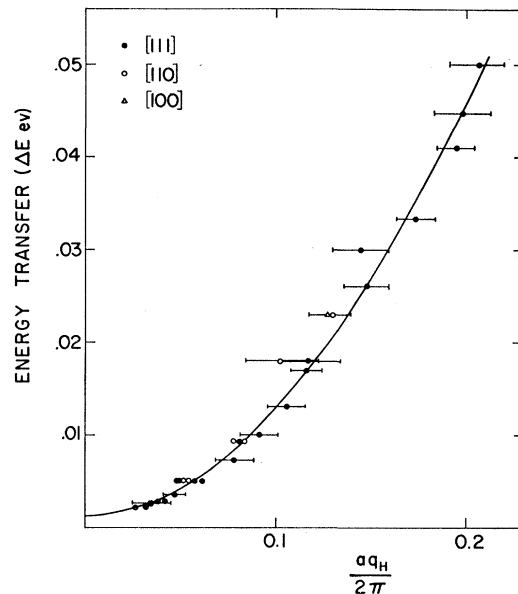


FIG. 2. The energy transfer  $\Delta E$  as a function of the reduced vector of the spin waves. The error bars are about half of the full width at half maximum of the neutron groups. The solid line is the best fit to Eq. (3).

ment with a value from the estimated elastic constants of the alloy.

The majority of the distributions were taken in the transverse  $[1,1,1]$  direction through the  $(1,1,1)$  reciprocal lattice point because the low velocity of sound in this direction leads to the largest separation in momenta of spin waves and phonons of the same energy, and also because the magnetic form factor is favorable.

## RESULTS

The energies and wave vectors of those neutron groups which were definitely established to be of spin-wave origin are shown in the dispersion curve (Fig. 2). The decrease of the peak intensity with the increase of wave vector, due to the limited spectrum of neutron energies which was available from the reactor combined with the population factor in the cross section, prohibited the extension of the curve beyond a value of the reduced vector of about 0.2. This represents a quarter of the distance to the zone boundary in the  $[1,1,1]$  direction. The values of the reduced vector were measured in the horizontal  $(0,1,1)$  plane, without consideration for the vertical divergence of the beam—this will be discussed later.

The errors have been estimated after consideration of the widths of the neutron groups and of the probable error in the determinations of  $\phi$  and  $\psi$ . The errors in  $\psi$  are partially eliminated from the dispersion curve since points were taken on either side of the  $(1,1,1)$  reciprocal lattice point. Errors in  $\phi$  were usually not so important because of the near isotropy of the spin-wave dispersion curve.

Figure 2 also shows points which were taken in the  $[0,1,1]$  and  $[1,0,0]$  direction of the reduced zone. The accuracy achieved in this experiment was not sufficient to demonstrate the dependence upon direction over the limited range of  $\mathbf{q}$  which was attainable, but approximate isotropy is observed to hold.

The field dependence of the intensities of neutron groups that represented spin waves with  $\hbar\omega \leq 0.003$  ev was not symmetrical about the reciprocal lattice point. On one side the effect was in approximate agreement with the theoretical prediction while on the other its magnitude was reduced by at least 50%. At present this anomaly is not understood.

### DISCUSSION

The dispersion law for a ferromagnetic cubic lattice of spins with nearest neighbor interaction only may be written<sup>9</sup>

$$\hbar\omega = C + 2JS[Z - \sum_l \cos(\mathbf{q} \cdot \mathbf{l})], \quad (2)$$

where the summation is over the  $Z$  vectors denoted by  $\mathbf{l}$  which join the central atom to its  $Z$  nearest neighbors. The constant term  $C$  allows for the presence of the external and anisotropy fields and, using the relations of Herring and Kittel,<sup>10</sup> has a value of roughly  $10^{-4}$  ev for the cobalt alloy in the geometry of this experiment.

For the  $[1,1,1]$  direction of the face-centered cubic alloy this expression reduces to

$$\hbar\omega = C + 12JS \left[ 1 - \cos\left(\frac{2\pi}{\sqrt{3}} \cdot \frac{qa}{2\pi}\right) \right], \quad (3)$$

where  $a$  is the lattice constant.

The continuous line in Fig. 2 represents a least squares fit of Eq. (3) to the experimental points and yields a value of  $JS = (1.47 \pm 0.15) \times 10^{-2}$  ev and an apparent value of  $C = (1.3 \pm 0.5) \times 10^{-3}$  ev. However, in this experiment the values of the wave vector were measured in the horizontal plane and allowance must be made for the vertical divergence of the neutron beams which tend to increase the effective value of the wave vector. The mean divergence is estimated, from the geometry of the spectrometer, to be  $1.5 \pm 0.3^\circ$ .

For small values of  $\mathbf{q}$ , neglecting fourth and higher

order terms, (3) may be written

$$\hbar\omega = C + 12JS \frac{4\pi^2}{3} \left( \frac{aq_v}{2\pi} \right)^2 + 12JS \frac{4\pi^2}{3} \left( \frac{aq_H}{2\pi} \right)^2, \quad (4)$$

where  $q_H$  and  $q_v$  represent the components of the effective wave vector measured along and perpendicular to the horizontal plane. As a good approximation the value of  $aq_v/2\pi$  may be considered as a constant of magnitude  $0.04 \pm 0.01$  over the range of measurements. Hence the measured value of  $C$  contained a contribution from the second term on the right-hand side of (4). The magnitude of this term is  $(1.6 \pm 0.6) \times 10^{-3}$  ev and this represents the principal contribution to the measured value of  $C$ , thus masking the effect of the applied and anisotropy fields. From this experiment all that can be said is that  $C < 10^{-3}$  ev.

Because of the nonintegral number of Bohr magnetons there are difficulties in obtaining the value of the exchange integral  $J$ , and in comparing it with values obtained from other work. If  $S$  is simply taken to be  $p_B/2 = 0.92$ , then the determined value of  $J$  is  $1.60 \times 10^{-2}$  ev, and is to be compared with a value  $J = 2.42 \times 10^{-2}$  ev similarly derived from the spin-wave interpretation of the low-temperature magnetization data of Weiss and Forrer.<sup>7</sup> Brown and Luttinger<sup>11</sup> have collected values of  $kT_c/J$  as predicted by the Kramers-Opechowski method for a fcc lattice of spins. They also give a semiclassical method for the generalization of the ratios to arbitrary  $S$ . This procedure, when adopted for this alloy, results in a value of  $J = 1.12 \times 10^{-2}$  ev.

Because of the limited range of measurements, the experimental curve is not very sensitive to the range of interaction between spins.

Study of the widths of the neutron groups is difficult because of the comparatively poor resolution. Considering the full width at half maximum of the groups ( $W$ ), a limit on the mean lifetime ( $\tau_0$ ) may be set by using the relation  $W\tau_0 = \hbar$ . The only conclusion that may be reached is that the lifetimes represented by the sharpest neutron groups are longer than  $3 \times 10^{-13}$  sec.

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<sup>9</sup> J. Van Kranendonk and J. H. Van Vleck, *Revs. Modern Phys.* **30**, 1 (1958).

<sup>10</sup> C. Herring and C. Kittel, *Phys. Rev.* **81**, 869 (1951).

<sup>11</sup> H. A. Brown and J. M. Luttinger, *Phys. Rev.* **100**, 685 (1955).