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Direct Observation of the Angular Distribution of Neutrons Scattered at Small Angles by Spin Waves in Fe–Ni Alloys

S. A. WERNER
Scientific Research Staff, Ford Motor Company, Dearborn, Michigan and *Department of Nuclear Engineering, University of Michigan, Ann Arbor, Michigan*
E. WIENER (AVNEAR)* AND E. GÜRMEN
Department of Nuclear Engineering, University of Michigan, Ann Arbor, Michigan
AND
A. ARROTT
Scientific Research Staff, Ford Motor Company, Dearborn, Michigan and *Department of Physics, Simon Fraser University, Burnaby, British Columbia, Canada*

Extensive measurements of the small-angle scattering of neutrons by spin waves in Fe–Ni alloys using polycrystalline samples and a polyenergetic beam have been made by Hatherly *et al.* Their spin-wave stiffness D was found not to agree with later measurements using the diffraction method on $Fe_{0.5}Ni_{0.5}$ and $Fe_{0.2}Ni_{0.8}$ single crystals. In an effort to resolve this disagreement we employed a new technique to observe the small-angle scattering by spin waves in these materials that uses a double-Si-crystal spectrometer, monoenergetic neutrons, and single-crystal samples. Our result for D for the $Fe_{0.5}Ni_{0.5}$ sample agrees with the previous single-crystal data. However our data on $Fe_{0.2}Ni_{0.8}$ is in closer agreement with the earlier small-angle scattering measurements. The effect of the dipole–dipole interaction on the spin-wave energy has been clearly observed in $Fe_{0.5}Ni_{0.5}$. Two discontinuities are predicted and observed to occur in the scattering profiles. However, an additional discontinuity is observed in our $Fe_{0.7}Ni_{0.3}$ single crystal, which may indicate another scattering surface of unknown origin.

I. INTRODUCTION

Several experimental techniques have been used over the past few years to measure the spin-wave stiffness parameter D in Fe–Ni alloys. The large differences for $Fe_{0.2}Ni_{0.8}$ and $Fe_{0.5}Ni_{0.5}$ (at room temperature) between the small-angle scattering (SAS) results[1,2] and the diffraction method[3] (DM) results has raised some speculation that D may be sample-dependent.[3] The SAS data were taken on polycrystalline material and the DM data were obtained on a single crystal. Although there was an apparent agreement between the DM data on $Fe_{0.2}Ni_{0.8}$ and the spin-wave resonance method[4] for the value of D, the constant term in the dispersion law was quite different.

The data reported here were obtained using a new technique to observe the small-angle scattering of neutrons by spin waves. The same single-crystal samples ($Fe_{0.2}Ni_{0.8}$ and $Fe_{0.5}Ni_{0.5}$) used in the DM experiments were used in these experiments.[5]

II. EXPERIMENTAL TECHNIQUE

The techniques employed in these experiments uses two silicon crystals (mosaic spread ≈2.5 min) placed in the parallel position with the Fe–Ni crystal interposed between them.[6] The first Si crystal is the monochromator and the second is the analyzer (A) which is cooled to liquid-N_2 temperature to reduce the phonon background. The sample crystal is placed very close to the analyzer so that all the neutrons that are scattered at small angles by spin waves strike the analyzer. A "wide-open" detector is positioned in such a way that all the neutrons that are Bragg reflected by the analyzer are counted.

The "trajectory" of a given neutron in $\mathbf{k}$ space is shown in Fig. 1. A neutron of wave vector $\mathbf{k}_0$ incident on the monochromator is reflected by the (111) reciprocal-lattice vector $\mathbf{G}_M$ giving the wavevector $\mathbf{k}_1$ incident on the Fe–Ni sample. This neutron is scattered off a spin wave of wave vector $\mathbf{q}$ and gives rise to the wave vector $\mathbf{k}_2$ incident on the analyzer. Upon reflection by the analyzer scattering vector $\mathbf{G}_A$, the outgoing ray $\mathbf{k}_3$ is counted. There are, of course, other spin waves that will reflect the incident wave $\mathbf{k}_1$. The locus[7] of these $\mathbf{q}$ vectors is the scattering surface, which is simply two spheres of very small radii (for a quadratic dispersion law). For a given misset angle ϕ of the analyzer, all spin waves having their $\mathbf{q}$ vectors on the intersection of the plane *a-a* with the scattering surface give rise to observed intensity. We call *a-a* the "plane of integration" (PI). The PI is perpendicular to $\mathbf{G}_A$, and is parallel to the bisector O-A of $\mathbf{G}_A$. As the analyzer is rotated the bisector O–A rotates and consequently so does the PI. To an excellent approximation for small angles ϕ, the PI simply moves in a direction perpendicular to $\mathbf{G}_M$. We first pick up intensity from the creation surface as the PI moves upward in this diagram; subsequently, we obtain a jump in intensity when the PI touches the annihilation surface. We obtain intensity due to both the creation and annihilation of spin waves until the PI passes over the creation surface, at which point the intensity drops to a level appropriate to annihilation of spin waves only. The intensity goes to zero when the PI moves outside the annihilation surface.

The lack of perfect collimation has the effect of producing an array of scattering surfaces displaced from each other along the line $m'-m'$, and consequently has essentially no effect on the instrumental resolution. However, the mosaic spread of the monochromator has the effect of displacing the array of scattering surfaces perpendicularly to the PI, and does influence the resolution. In fact, the instrumental resolution is the folding together of the mosaic distributions of the monochromator and analyzer, which is simply a rocking curve (of the analyzer) with the sample removed. The absolute intensity of this rocking curve modified by the sample transmission is subtracted from the data (point by point) with the sample in place to give the small-angle scattering, which is found to be due almost entirely to spin waves.

III. RESULTS ON Fe–Ni

We show in Fig. 2(a) data obtained on $Fe_{0.5}Ni_{0.5}$ using this method in the presence of a magnetic field $H=3.5$ kG applied parallel and perpendicular to the horizontal plane. If there were no dipole-dipole effect, these two sets of data would have the same shape. The difference ($\Delta\phi\approx 0.02°$) in the cutoffs between these two sets of data agrees with that calculated from the Holstein–Primakoff theory of spin waves. From a calculation of the scattering surfaces using a magnetization $M=1350$ G, $H=3500$ G (and taking into account the effect of resolution), we find that these cutoffs require

$$D=(226\pm 10)\ \text{meV-Å}^2.$$

Similar data for the parallel field case on $Fe_{0.2}Ni_{0.8}$ are shown in Fig. 2(b). We find that for $M=835$ G, $H=3000$ G, these cutoffs require

$$D=(320\pm 15)\ \text{meV-Å}^2.$$

Our data on fcc $Fe_{0.7}Ni_{0.3}$ shows three jumps in the intensity on each side of $\phi=0$. The origin of the discontinuity is not yet understood. If we use the last cutoff angle, $M=435$ G, and $H=3000$ G, the stiffness (at room temperature) is

$$D=(70\pm 5)\ \text{meV-Å}^2.$$

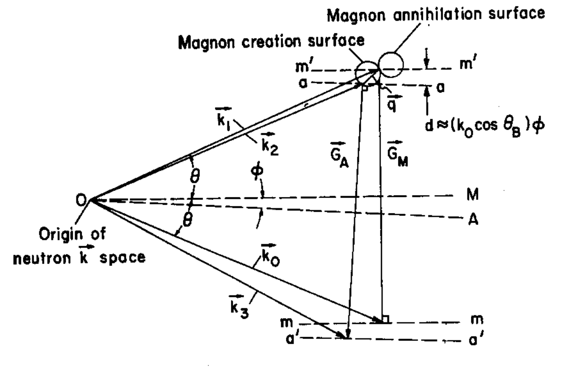
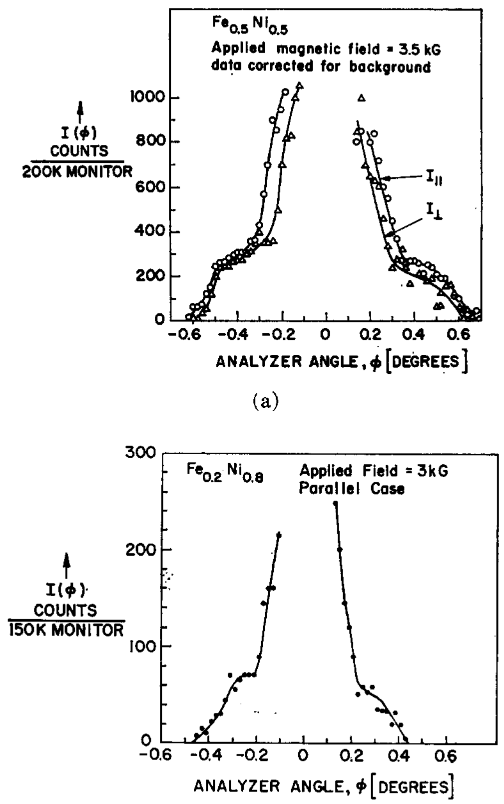


Fig. I

FIG. 1. Description in **k** space of the double-crystal technique for observing small-angle scattering from spin waves. In the experiments $k_0=3.48$ Å^{-1} and $\theta_B=17.2°$.



(a)

(b)

FIG. 2. (a) Intensity as a function of analyzer setting for small-angle spin-wave scattering in $Fe_{0.5}Ni_{0.5}$. (b) Intensity as a function of analyzer setting for small-angle spin-wave scattering in $Fe_{0.2}Ni_{0.8}$.

The spin-wave stiffness obtained for $Fe_{0.5}Ni_{0.5}$ is between the DM result (245±15 meV-Å^2) and the SAS value ($D\approx 210$ meV-Å^2) at room temperature. However, the limits of error overlap in both cases. Our measurements on $Fe_{0.2}Ni_{0.8}$ are in sharp disagreement with DM results $D=(252\pm 15$ meV-A$^2)$ on the same crystal. There is some confusion on the SAS result for D in this alloy quoted in Ref. 3, which gives $D=400$ meV-Å^2. Hatherly *et al.* give this value of D for low temperatures; their measurements at room temperature gave $D\approx 323$ meV-A.[8] Consequently, our measurement of the spin-wave stiffness in $Fe_{0.2}Ni_{0.8}$ is in very close agreement with the previous SAS measurements. The reason for the discrepancy between these two neutron techniques is not known.

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* On leave from Soreg Nuclear Research Center, Yavne, Israel.

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[5] We are grateful to G. Shirane at Brookhaven for the loan of these samples.

[6] This method resembles the experiments on slit diffraction of neutrons reported by C. G. Shull, Phys. Rev. **179,** 752 (1969).

[7] See Ref. 2 for a discussion of the scattering surfaces.

[8] M. W. Stringfellow (private communication). This value of D has not been corrected for magnetic field and dipole–dipole effects, which would decrease it by about 5%.

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Magnetic Excitations in a Cr–Mn Alloy by Inelastic Neutron Scattering

S. K. SINHA, S. H. LIU, AND N. WAKABAYASHI

Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa 50010

AND

L. D. MUHLESTEIN

Department of Physics, University of Missouri, Columbia, Missouri 65201

The inelastic magnetic scattering from a $Cr_{0.98}Mn_{0.02}$ alloy single crystal has been studied both below and above the Néel temperature. In the spin-wave regime at 300 K, we have obtained a value of 1.29×10^{7} cm/sec for the spin-wave velocity by using the diffraction method and suitably extracting the resolution function. At a temperature of 1.16 T_N, we have observed quite strong inelastic magnetic scattering and fitted the results quite well by calculating the imaginary part of $\chi(Q)$ for paramagnetic chromium. A triple-axis spectrometer was used to study the magnetic excitations in a crystal of $Cr_{0.99}Mn_{0.01}$ below, above and close to the Néel temperature of 514 K. At low temperatures, peaks at the magnetic superlattice points were seen consistent with the above spin-wave velocities. These appear to go smoothly into the slightly broader paramagnon peaks as the temperature was raised through T_N without any sudden change of intensity. The absence of Stoner excitations near T_N suggests that either the neutron cross section for these is very low or else that the sample is gapless near T_N due to phonon broadening of the electronic states. Full details are published elsewhere.[1]

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[1] S. K. Sinha, S. H. Liu, N. Wakabayashi, and L. D. Muhlestein, Phys. Rev. Lett. **23,** 311 (1969); Phys. Rev. (to be published).

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Short-Range Order Effects in Paramagnetic Neutron Scattering

R. TAHIR-KHELI*

Temple University, Philadelphia, Pennsylvania

AND

F. MALINOSKI

Frankford Arsenal, Philadelphia, Pennsylvania and *Temple University, Philadelphia, Pennsylvania*

To study the effects of short-range order on paramagnetic lineshapes, we have computed temperature-dependent frequency moments for an exchange-coupled paramagnet with uniaxial, single-ion anisotropy, i.e.,

$$\mathcal{H}=-\sum_{f_1,f_2} I(f_1,f_2)\,\mathbf{S}_{f_1}\cdot\mathbf{S}_{f_2}-D\sum_{f}(S_f^{z})^2.$$

We have used phenomenological procedures to construct the frequency and wave-vector-dependent Van Hove correlation function. The results are discussed with reference to the inelastic scattering cross section of slow neutrons from such paramagnetic systems.

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