Neutron-Scattering Measurement of the Spin-Wave Spectra for Nickel

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Improvements in neutron spectroscopy have made possible measurements of the spin-wave spectra for nickel over much of the zone for the [111] and [100] directions. Broadening of the spin-wave excitations has been directly observed for the [111] direction and the nature of the optical spin wave has been further characterized for the [100] direction. Comparison of the measured results is made with recent theoretical calculations.

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The magnetism of nickel continues to be the subject of many theoretical and experimental investigations. Since the magnetic electrons in nickel appear to have properties that fit both a localized and an itinerant picture depending on the experiment, an understanding of the magnetism of nickel is one of the most difficult problems in condensed-matter physics. A measurement of the complete spin-wave spectra provides detailed information that serves as an excellent test of any theoretical calculation made in the hopes of understanding the magnetic interactions in a material. Measuring the spin-wave spectra for nickel is a very difficult neutron-scattering experiment since the spin-wave energies are much larger than the energies of thermal neutrons. However, by use of a large sample of ^{60}Ni and the improved triple-axis spectrometer located at the hot source of the Institut Laue-Langevin reactor we were able to obtain a nearly complete measurement of the spin-wave dispersion curves for the [111] and [100] directions. In this Letter these measurements are presented and the results are compared with the recent band calculations by Cooke, Blackman, and Morgan[1] and calculations made by Callaway *et al.*[2] using density-functional theory in the local-density approximation.

As neutron-scattering techniques have improved, a larger part of the spin-wave spectra for nickel has been established, and Mook and Tocchetti[3] and references therein discuss previous measurements for nickel. The present measurements were made at the same beam position as those discussed in Ref. 3 but the spectrometer has been greatly improved. The biggest improvement stems from the very large monochromator and analyzer assemblies made from an array of large copper crystals that can be adjusted continously in vertical curvature for maximum neutron intensity. The sample was a high-quality 400-g ^{60}Ni crystal which gave clean scattering results free from incoherent scattering effects and highly reduced contributions from phonon scattering. Resonance filters of Er, In, and Hf were used to avoid half-wavelength contamination and Sm filters removed unwanted low-energy neutron background. The high spin-wave energy of nickel and the necessity of keeping the momentum transfer Q small so that the magnetic form factor remains at a reasonable value required the use of high-energy neutrons. In order to carry out the measurements for the highest spin-wave energies, neutron energies of about 200 THz (1 THz $\cong$4.14 meV) were needed. Fortunately the hot source provided a reasonable flux of neutrons in this energy range.

The data were analyzed by calculation of the intensity expected from the spectrometer by means of a four-dimensional integration in which the resolution function of the spectrometer was convoluted with a dispersion surface of the form

$$\sum_{i=1}^{4}\sum_{j=1}^{4} X_i B_{ij} X_j + T_j X_i + H = 0, \tag{1}$$

where B is a 4×4 symmetric matrix, T is a four-component vector, and H is a constant. This is a general four-dimensional quadratic equation in momentum-energy space and represents the dispersion surface of interest. The resolution function was calculated from the known crystal mosaic spreads and spectrometer collimations. The four-dimensional integration was performed by a technique suggested by Haywood[4] and a least-squares comparison was made between the dispersion parameters in (1), an intrinsic spin-wave width assumed to be Gaussian, and an overall height scale factor. Adjustment of the parameters was made until a good quality-of-fit parameter χ was obtained. In all cases an excellent representation of the data was obtained with this process.

Figure 1 shows the dispersion curve for the [111] direction obtained by the above procedure. It is found that single-particle excitations or Stoner modes do not completely damp out the spin-wave mode and that the spin wave continues to propogate into the Stoner con-

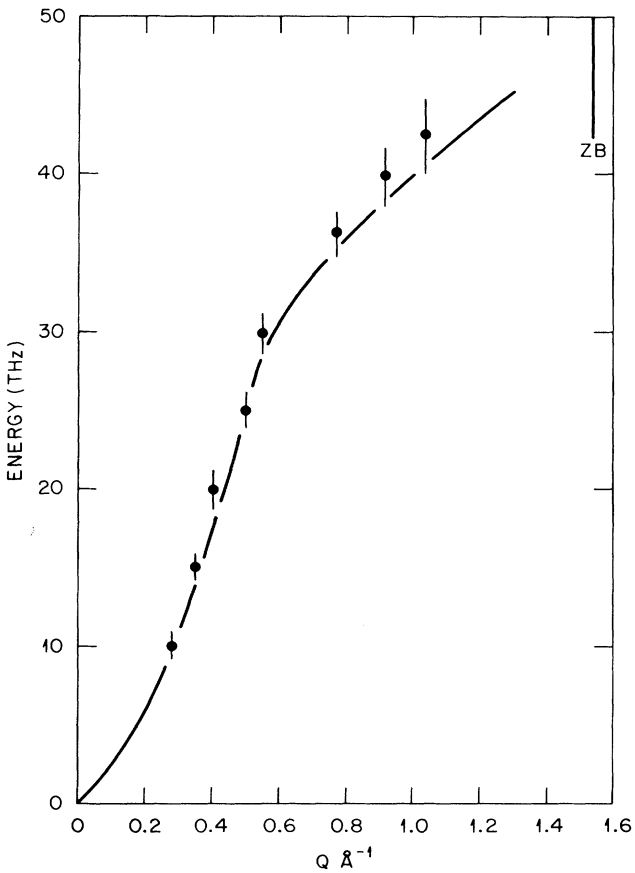

FIG. 1. Spin-wave dispersion curve for the [111] direction. The solid curve is from the calculation in Ref. 1. ZB shows the position of the zone boundary.

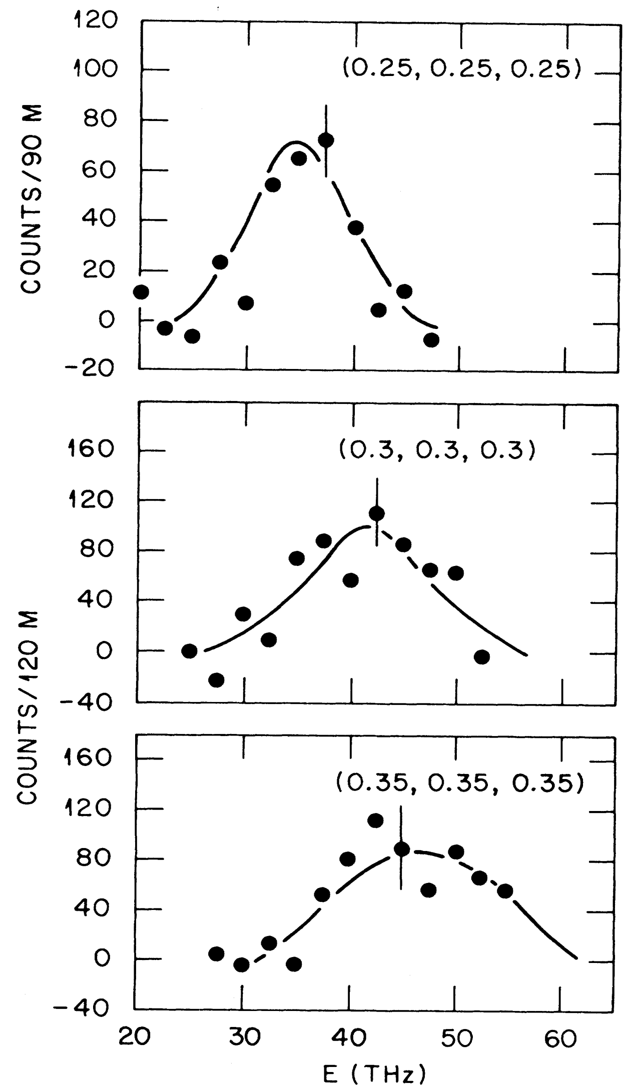

FIG. 2. Constant-Q scans of spin waves for the [111] direction. The solid line is a least-squares fit of the convolution of the spectrometer resolution and a dispersion surface represented by Eq. (1). The data shown are the result of subtracting a background scan with the sample removed from the measured result. The counting time was determined from a monitor detector to account for any small variations in reactor power, and averaged about 20 min a point.

tinuum, but with a much reduced intensity. The solid line in Fig. 1 is from the Cooke, Blackman and Morgan[1] calculation and the agreement with experiment is very good. The calculation of Callaway *et al.*[2] produces a similar result. The measurements were made at room temperature while the theory is for zero temperature; however, we do not expect this to make very much difference for these high energies. The calculations show that the spin wave broadens and decreases in intensity as the energy increases. We were able to measure directly the broadening of the spin wave as it extended into the Stoner continuum at high energies. Figure 2 shows three constant-Q scans made near the top of the [111] dispersion curve. The Q position of the scan is given in zone-boundary units referenced to the [111] reciprocal-lattice point. The solid lines are the results of the least-squares fits of the convolution of the dispersion surface and the resolution function as outlined above. The result at [0.25 0.25 0.25] was no wider than the resolution employed in the experiment and the least-squares fit gave no excess width to the spin wave. However, excess widths of 5 ± 2 and 8 ± 4 THz were required in order to fit the results at [0.30 0.30 0.30] and [0.35 0.35 0.35], respectively. This is the first direct measurement of the spin-wave broadening as the spin wave encounters a dense region of Stoner excitations. The measurements were made at a constant final energy of 87.17 THz so that half-wavelength contamination could be eliminated by an indium resonance filter. Despite the high energy used the scattering triangle could not be closed for energies above 55 THz which made completion of the [0.35 0.35 0.35] scan impossible. Nevertheless, the expected broadening of the spin waves is clearly observed. The broadening is in good agreement with the calculation of Cooke, Blackman, and Morgan[1] but Callaway *et al.*[2] give results that are much too broad in energy. An absolute intensity of the spin-wave excitations normally can be obtained from the constant-Q scans by comparison to a phonon measurement. However, a phonon could not be clearly measured with the broad resolutions needed for the high-energy measurements so that a direct comparison could not be made. A knowledge of the efficiency of the analyzing crystal at various wavelengths was thus required for an absolute cross-section determination. With use of the best available information a cross section of about 3 mb was determined for the [0.25 0.25 0.25] spin wave.

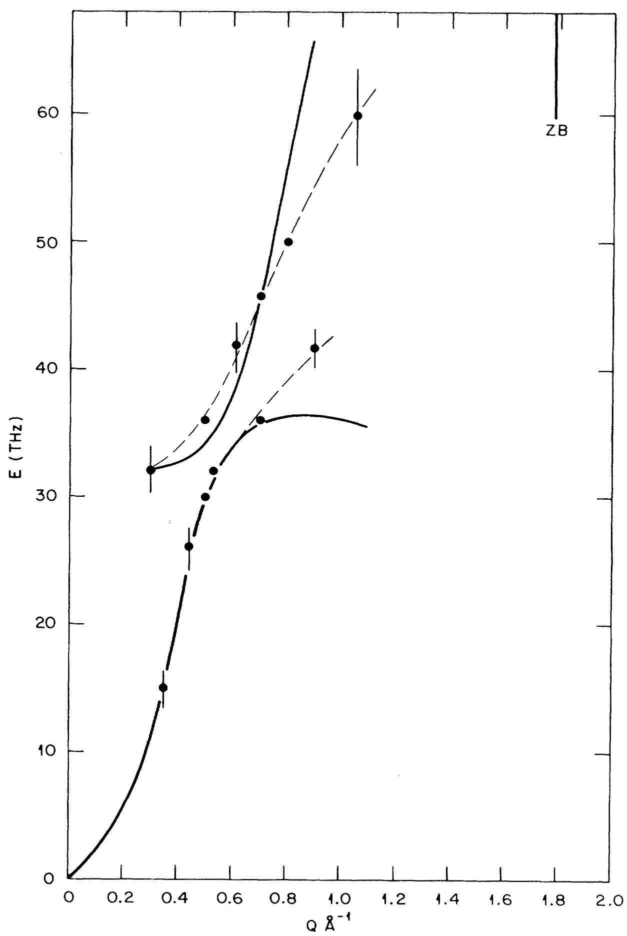

FIG. 3. Spin-wave dispersion curve for the [100] direction. The solid line is from the calculation of Ref. 1. While the calculation appears to give a good qualitative representation of the data, notable departures are observed for the larger Q values.

This is an order of magnitude lower than that obtained by Callaway *et al.*[2] and while the measured result is not very accurate it does not substantiate the calculation. The calculation of Ref. 1 does not provide absolute intensity results.

Figure 3 shows the spin-wave dispersion curve for the [100] direction. The unusual structure of the dispersion curve for this direction between 30 and 40 THz was established earlier[3] and is in reasonable agreement with the calculation of Ref. 1 shown by the solid lines for about the first half of the zone. The calculation shows that an optical spin wave crosses the main spin-wave branch splitting it into two pieces. With the increased intensity available in this experiment we could follow the lower curve farther out into the zone and we find that it continues upward in energy and diminishes quickly in intensity. This disagrees with the calculation of Ref. 1 which shows it extending into the zone at a rather constant energy. The upper part of the curve seems to be in good agreement with calculation of Ref. 1 except for the higher energies. The calculation of Callaway *et al.*[2] appears to be incorrect for the [100] direction as no optical mode is predicted and the energy scale is much too low. The intensity of the scattering for the [100] direction near the optical-mode crossing was sufficient that higher resolution could be used than for the [111] direction. However, it would be desirable to make measurements with still better resolution to further understand the unusual phenomena in this region.

In conclusion the present measurements show that while the spin waves for nickel are strongly damped in the presence of Stoner modes, they remain measurable over much of the zone. Theoretical calculations are in good agreement with the position of the [111] branch but the local-density approximation calculations appear to give the widths incorrectly and the total cross section appears too high. The unusual behavior in the [100] direction is not given correctly by theoretical calculations although the result by Cooke, Blackman, and Morgan[1] appears to show many of the general features that are observed. It is hoped that the new measurements will serve as a guide for improving the theoretical results.

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