New Interpretation of Spin-Wave Behavior in Nickel

J. F. Cooke
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

and

J. A. Blackman and T. Morgan
Physics Department, University of Reading, Reading, England
(Received 29 August 1984)

Results from recent calculations of the neutron-scattering cross section for energy transfer up to 1 eV have led to a new picture of spin waves in nickel, one in which spin waves exist out to the zone boundary. Our prediction of the dispersion curve and peak widths is in general agreement with data from subsequent neutron-scattering experiments. This provides additional evidence that the "average" spin-splitting energy for nickel is in the 300- to 400-meV range.

PACS numbers: 75.30.Ds, 61.12.Fy

Past inelastic neutron-scattering experiments have provided considerable information about the unusual properties of spin waves in the $3d$ transition-metal ferromagnets nickel[1] and iron. Such information is useful to test theoretical models of ferromagnetism for metallic systems. Results from these experiments have supported the itinerant-electron theory but did not provide a complete description of spin waves in these materials. This is because neutron-scattering cross sections with energy transfer above 150 meV are very difficult, if not impossible, to measure with conventional steady-state reactors. Because of this, and the need to reduce computer time to a minimum, early theoretical calculations of the inelastic neutron-scattering cross section, magnetic susceptibility, and spin-wave behavior were restricted to energies below 200 meV.[2] These calculations based on "realistic" energy bands were found to be in excellent agreement with experiment for energies below 100 meV. Above 100 meV, these calculations predicted the existence of an "optical" spin-wave mode at about 130–140 meV along the [100] direction in nickel and a spin-wave branch which continued above the 200-meV cutoff. Neither of these effects was found in the early experiments. Subsequent experiments using the hot source at the Institut Laue-Langevin (ILL) reactor confirmed the existence of the "optic" mode at the predicted energy[3] but were unable to provide information about the higher-energy part of the dispersion curve.

Because of the recent improvement of the hot source at the ILL and the development of neutron spallation sources, which should be useful for energies up to 800 meV, we decided to map out the complete theoretical spin-wave spectra for nickel and iron. In this Letter we present some results for nickel.

The total magnetic inelastic cross section consists of a transverse part, which contains information about spin-flip scattering, and a longitudinal part corresponding to spin-nonflip scattering. The spin-polarized electronic wave function is expanded as

$$\psi_{nk\sigma}(\mathbf{r}) = \textstyle\sum_{\mu} a_{n\mu\sigma}(k)\phi_{\mu}^{\sigma}(r), \tag{1}$$

where n and $\mathbf{k}$ are band and wave-vector labels, respectively, $\{\phi_{\mu}^{\sigma}(r)\}$ are symmetry orbitals, $\{a_{n\mu\sigma}(k)\}$ are corresponding expansion coefficients, and μ is a symmetry label which runs over s, p, and d symmetry terms. Then the approximate, random-phase approximation expression for the transverse part of the cross section has the form

$$I(\mathbf{Q},\omega) = F_d(\mathbf{Q}) \lim_{\epsilon\to 0} \mathrm{Im}\left\{\sum_{\substack{\mu\nu\\ \eta}}[I+\Gamma(\mathbf{q},z)]_{\mu\nu}^{-1}\Gamma_{\nu\eta}(q,z)/U_{\eta}\right\}_{z=\omega+i\epsilon}, \tag{2}$$

$$\Gamma_{\mu\nu}(\mathbf{q},z) = \frac{U_\nu}{N}\sum_{\substack{nm\\ \mathbf{k}}}\frac{f_{n\mathbf{k}\uparrow}-f_{m\mathbf{k}+\mathbf{q}\downarrow}}{z-E(m\mathbf{k}+\mathbf{q}\downarrow)+E(n\mathbf{k}\uparrow)} a_{n\mu\uparrow}(\mathbf{k})a_{m\mu\downarrow}(\mathbf{k}+\mathbf{q})a_{n\nu\uparrow}(\mathbf{k})a_{m\nu\downarrow}(\mathbf{k}+\mathbf{q}), \tag{3}$$

where $\mathbf{Q}=\mathbf{G}+\mathbf{q}$, $\mathbf{G}$ is a reciprocal lattice vector, and $\mathbf{q}$ is restricted to the first Brillouin zone, $E(nk\sigma)$ is the electronic energy, $f_{nk\sigma}$ is the Fermi occupation number, and the $\{U_\nu\}$ are parameters determined from the band structure.[2] The symmetry sums in Eq. (2) are restricted to d terms in our calculations. There is a similar expression for the longitudinal cross section.

Notice that $I(\mathbf{Q},\omega)$ is completely determined from the band structure, i.e., there are no adjustable parameters. We have evaluated it for a "realistic" band structure used in previous calculations for energy transfer below 200 meV.[2] The Brillouin-zone sums in Eq. (3) were performed using the tetrahedron method.

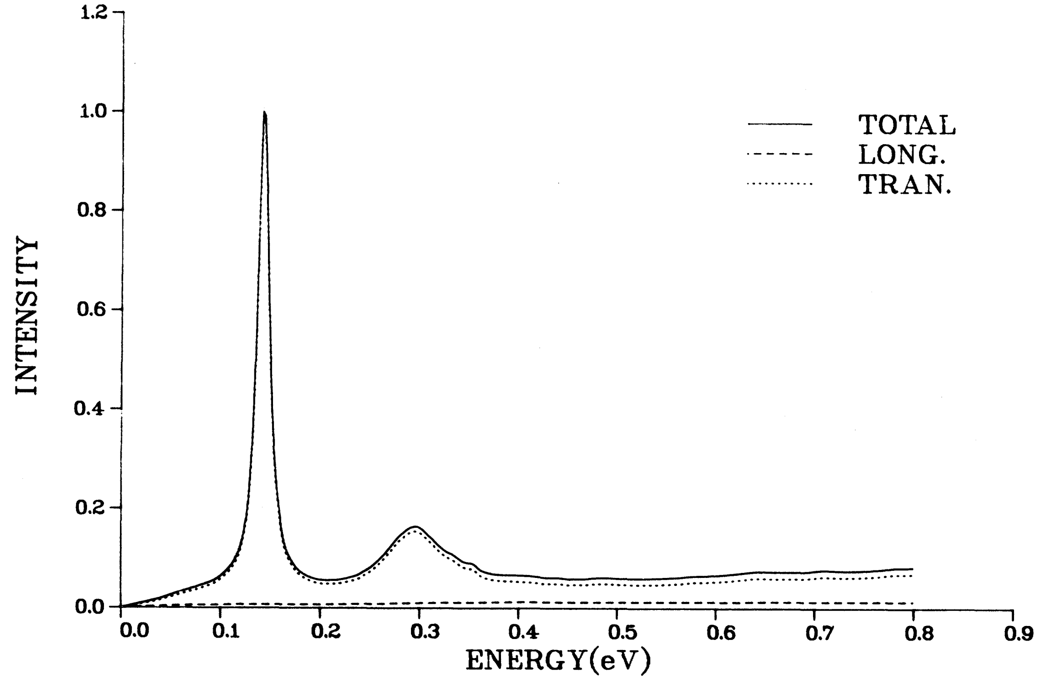

FIG. 1. Magnetic neutron-scattering cross section for $\mathbf{q} = (0.5625, 0.0, 0.0) \times (2\pi/a_0)$ for nickel.

The calculations of $I(\mathbf{Q}, \omega)$ were carried out for fixed $\mathbf{q}$ (constant $\mathbf{q}$) as a function of ω for energies up to 1 eV. Results were obtained for $\mathbf{q}$ along [100] and [111] on a mesh corresponding to $\Delta q = 0.0625$ (units of $2\pi/a_0$). The spin waves show up as peaks in the cross section. The positions of the peaks determine the spin-wave energy and the peak widths reflect inverse lifetimes. An example of this type of result is shown in Fig. 1. A summary of results from an extensive series of calculations for nickel is given in Figs. 2 and 3. The two main features of the [100] branch (Fig. 2) are the apparent crossing of an acoustic mode and "optic" mode and the continuation of the upper branch to about 350 meV. The spin-wave peak is very sharp for energies below 100 meV. The peak width broadens somewhat and the peak height drops by about an order

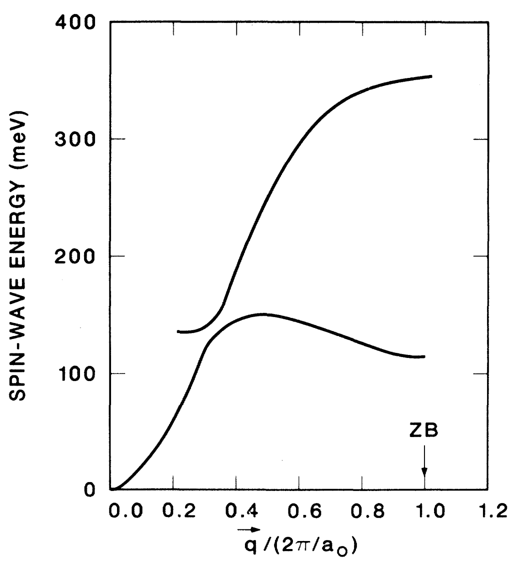

FIG. 2. Spin-wave dispersion curve along [100] for nickel.

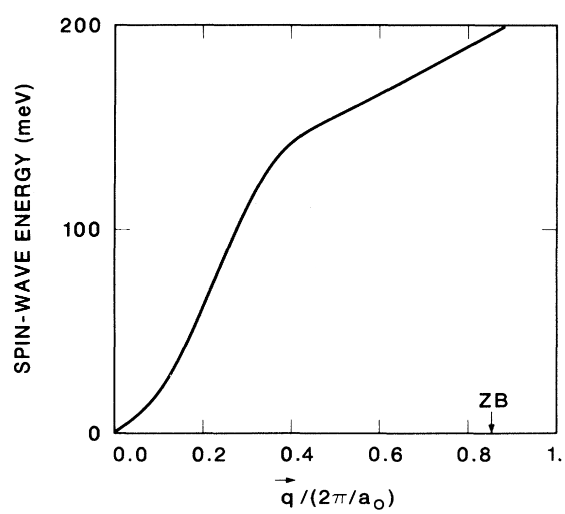

FIG. 3. Spin-wave dispersion curve along [111] for nickel.

of magnitude in the region where the lower branch bends over. Relatively small peaks associated with this lower branch persist out to the zone boundary. The peak width of the upper branch increases rather slowly at first but broadens significantly as the zone boundary is approached. The results in Fig. 3 are for **q** along [111], where no "optic" branch is found. The spin-wave peaks broaden considerably and the intensity drops significantly as the zone boundary is approached.

These numerical calculations provide a more coherent theoretical picture of spin-wave behavior in nickel. Early theoretical work had established that spin waves might run into a continuum of single-particle spin-flip excitations (Stoner excitations) and disappear at some wave vector inside the first Brillouin zone. Early experimental work on nickel found that spin-wave intensities did drop dramatically to undetectable limits at about one-quarter of the way to the zone boundary.[1] The energy of the spin wave at this point was about 100 meV. This was taken as direct evidence of the spin waves disappearing into the Stoner continuum. The problem with this interpretation was that the energy characteristic of Stoner excitations at such relatively small wave vectors should be near the spin-splitting energy, which at that time was estimated to be in the range 600 to 800 meV.

The results given in Figs. 2 and 3 provide a straightforward resolution of this conflict. The "disappearance" of the spin waves along [100] in the early experiments was associated with the crossing of the "optic" and acoustic branches, a phenomena which was totally unexpected at the time. Our calculations indicate, however, that small remnants of spin-wave peaks exist out to the zone boundary in both the [100] and [111] directions. We interpret the substantial broadening of the spin-wave peaks in the upper branch along [100] near the zone boundary to be associated with the relatively large density of Stoner excitations which is known to occur around the "average" spin-splitting energy (~ 400 meV for our band structure). It should be noted that because of *s-p-d* hybridization effects, Stoner excitations occur at essentially all energies, a fact which is reflected in the finite widths of the spin-wave peaks for all nonzero energies.

Results from recent neutron-scattering experiments carried out at the ILL by Mook and Paul[4] subsequent to our work appear to be in general agreement with the theoretical behavior. There is, however, a problem with the lower-energy branch along [100]. As can be seen from Fig. 1, the peak associated with the lower branch is larger and sharper than the one for the upper branch. This situation persists out to the zone boundary. The neutron measurements of Mook and Paul[4] were able to detect the upper branch near the zone boundary but not the lower one. This inconsistency might be attributed to neglect of all but the pure *d*-symmetry terms in the evaluation of the cross-section expression given in Eq. (2). We do not expect that the inclusion of the *s*, *p*, and cross-symmetry terms will significantly affect the position of the peaks but it could alter the peak widths, especially at lower energies. Work is currently under way to generalize our computer programs to include these terms.

The general form of spin-wave spectra shown in Figs. 2 and 3 appears to be a general feature of our theory and does not depend on details of the band structure. However, our results for the spin-wave spectra above 100 meV differ significantly from those found by Callaway *et al.*[5] Their results for the spin-wave energy, based on local density theory, are similar to ours for **q** along [111] but, as can be seen from Table I, our prediction of peak widths (inverse lifetimes) are significantly smaller and closer to experiment. For **q** along [100], their results for the spin-wave energy and peak width also differ from ours; in particular, no "optic" mode is predicted. It is not clear at present whether these differences result from theory or from numerical procedures used to evaluate the complicated cross-section expressions.

In summary, our calculations indicate that for nickel the spin-wave peaks persist throughout most of the Brillouin zone with intensities that decrease significantly as the zone boundary is approached. We associate the very broad peaks found in the theoretical calculations and in the neutron experiments near the [100] zone boundary with the spin waves running into the large density of Stoner excitations near the "average" spin-splitting energy, $\bar{\Delta}$. This means that $\bar{\Delta}$ is in the range 300 to 400 meV, a result which is consistent with angular-resolved photoemission results and a factor of 2 less than predicted by local-density theory. In conclusion, we would like to mention that very preliminary and somewhat crude calculations for iron indicate that, like in nickel, spin waves exist throughout most of the zone with "optic" mode(s) along [100]. The spin-wave energies near the zone boundary are

TABLE I. Spin-wave peak widths (full width at half maximum) in millielectronvolts for **q** along [111]. Blank spaces indicate no available data.

$\lvert \mathbf{q} \rvert$ ($\mathrm{\AA}^{-1}$)	Mook and Paul (Ref. 4) (Expt.)	Cooke *et al.*	Callaway *et al.* (Ref. 5)
0.19		4	
0.26			13
0.52		16	44
0.77		16	54
0.92	21 ± 8		
1.1	33 ± 17	35	

much higher than in nickel. A more detailed description of our results for nickel and results for iron will be published in the near future.

This research was sponsored by the Division of Materials Sciences, U. S. Department of Energy under Contract No. DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc.

[1]H. A. Mook, R. M. Nicklow, E. D. Thompson, and M. K. Wilkinson, J. Appl. Phys. **40**, 1450 (1969); H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. **30**, 556 (1973), and in *Proceedings of the Nineteenth Conference on Magnetism and Magnetic Materials, Boston—1973,* edited by C. D. Graham, Jr., and J. J. Rhyne, AIP Conference Proceedings No. 18 (American Institute of Physics, New York, 1974), p. 781.

[2]J. F. Cooke and H. L. Davis, in *Proceedings of the Eighteenth Conference on Magnetism and Magnetic Materials, Denver—1972,* edited by C. D. Graham, Jr., and J. J. Rhyne, AIP Conference Proceedings No. 10 (American Institute of Physics, New York, 1975), p. 1218; J. F. Cooke, Phys. Rev. B **7**, 1108 (1973); J. F. Cooke, J. W. Lynn, and H. L. Davis, Phys. Rev. B **21**, 4118 (1980). The bands used in the calculation are those used to generate the curve labeled CD2 in Fig. 9 of the previous reference.

[3]H. A. Mook and D. Tocchetti, Phys. Rev. Lett. **43**, 2027 (1979).

[4]H. A. Mook and D. Mck. Paul, to be published.

[5]Joseph Callaway, Arun K. Chatterjee, Sat. P. Singhel, and A. Zeigler, Phys. Rev. B **28**, 3818 (1983).