Prediction of High-Energy Spin-Wave Excitation in Iron

J. A. Blackman and T. Morgan
Physics Department, University of Reading, Reading, England

and

J. F. Cooke
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831
(Received 31 July 1985)

Calculations have been made of inelastic-neutron-scattering cross sections for ferromagnetic iron. Quite unexpectedly, a rich picture of high-energy collective excitations has emerged. In particular, "optic" spin-waves modes with energies ranging to about 750 meV are predicted. Thus, iron is an ideal candidate for a novel high-energy-transfer experiment on recently developed high-energy neutron sources.

PACS numbers: 75.30.Ds, 61.12.−q, 75.10.Lp

Itinerant-electron theory has been very successful in giving a description of the low-temperature spin waves in the $3d$ transition-metal ferromagnets nickel and iron. Recent predictions[1,2] of distinct high-energy collective excitations above 100 meV in nickel were subsequently confirmed[3,4] by inelastic neutron scattering using the hot source at the Institut Laue-Langevin. Some discrepancies in detail exist between theory and experiment but, in broad terms, they are in agreement. In this Letter we report results of calculations for iron, an itinerant-electron ferromagnet with spin-splitting energy $\cong 2$ eV, some 5 times the value in nickel. From these calculations, we predict the existence of new collective excitations in Fe which exist both throughout the Brillouin zone and to energies much higher than were previously thought possible.

The theory on which the calculations are based is summarized as follows. The spin-polarized electronic wave functions are expanded as

$$\psi_{n\mathbf{k}\sigma}(\mathbf{r}) = \sum_{\mu} a_{n\mu\sigma}(\mathbf{k})\phi_{\mu}(\mathbf{r}), \qquad (1)$$

where n, $\mathbf{k}$, and σ are band, wave-vector, and spin labels, respectively. The $\{\phi_\mu(\mathbf{r})\}$ are symmetry orbitals with μ a symmetry label which runs over the nine (s,p,d) symmetry terms. The $\{a_{n\mu\sigma}(\mathbf{k})\}$ are the corresponding expansion coefficients. Within the random-phase approximation the transverse part of the magnetic inelastic cross section can be written as

$$\chi(\mathbf{Q},E) \sim \sum_{\mu\nu\eta\xi} F_{\mu\nu}(\mathbf{Q}) \left\{ \lim_{\epsilon\to 0} \mathrm{Im}([I+\Gamma(\mathbf{q},z)W]^{-1}\Gamma(\mathbf{q},z))_{z-E+i\epsilon} \right\}_{\mu\nu\eta\xi} F_{\xi\eta}(\mathbf{Q}), \qquad (2)$$

where F is a form factor, and $\mathbf{Q}=\mathbf{G}+\mathbf{q}$ with $\mathbf{G}$ a reciprocal-lattice vector and $\mathbf{q}$ restricted to the first Brillouin zone. The sum is taken only over the d-symmetry terms which are the dominant ones. The Γ matrix is defined by

$$\Gamma_{\mu\nu\eta\xi}(\mathbf{q},z) = \frac{1}{N}\sum_{nm\mathbf{k}} \frac{a_{n\mu\downarrow}(\mathbf{k})a_{m\nu\uparrow}(\mathbf{k}+\mathbf{q})a_{n\eta\downarrow}(\mathbf{k})a_{m\epsilon\uparrow}(\mathbf{k}+\mathbf{q})}{Z-E(m,\mathbf{k}+\mathbf{q},\uparrow)+E(n,\mathbf{k},\downarrow)}(f_{n\mathbf{k}\downarrow}-f_{m\,\mathbf{k}+\mathbf{q}\uparrow}), \qquad (3)$$

where $f_{n\mathbf{k}\sigma}$ are the Fermi occupation numbers and the $W_{\mu\nu\eta\xi}$ are matrix elements of a self-consistently screened electron-electron interaction:

$$W_{\mu\nu\eta\xi} = \int \phi_\mu(\mathbf{r})\phi_\nu(\mathbf{r}')U_{\mathrm{sc}}(\mathbf{r},\mathbf{r}')\phi_\eta(\mathbf{r}')\phi_\xi(\mathbf{r})\,d^3r\,d^3r'. \qquad (4)$$

The expression for the electronic energy within the random-phase approximation is then

$$E(n\mathbf{k}\sigma) = \epsilon(n\mathbf{k}\sigma) + \sum_{\eta\mu\nu} a_{n\mu\sigma}(\mathbf{k})a_{n\nu\sigma}(\mathbf{k})W_{\mu\eta\nu\eta}(N_\eta^\uparrow - N_\eta^\downarrow), \qquad (5)$$

$$N_\mu^\sigma = \frac{1}{N}\sum_{n\mathbf{k}} |a_{n\mu\sigma}(\mathbf{k})|^2 f_{n\mathbf{k}\sigma}, \qquad (6)$$

where N_μ^σ is the number of electrons of symmetry type μ and spin σ.

In past work the approximation has been made that W is diagonal (i.e., only $W_{\mu\mu\mu\mu}$ is nonzero), but the $W_{\mu\mu\mu\mu}$ were permitted to take on different values for the e_g and t_{2g} components. This allowed the development of a theory based on a two-parameter fit to static magnetic properties. The cross section as defined by Eqs. (2) and (3) is, however, not rigorously invariant with respect to point-group rotations with a diagonal W; i.e., results for q

along [100] and [001] might differ. Numerically, the discrepancy in nickel is minimal and does not affect previous results. In iron this effect is also minimal for the low-energy studies previously performed but, because of the strong e_g characteristics, it cannot be neglected in higher-energy studies.

There are several different, symmetrically correct, choices for the form of W, which can be characterized by a small number of parameters. The simplest is to include, in addition to the diagonal terms, elements of the type $W_{\mu\nu\nu\mu}$, with the condition $W_{\mu\nu\nu\mu} = W_{\mu\mu\mu\mu}$ for all ν, where μ and ν label orbitals belonging to the same irreducible representation. Because of the particular form of the rotation matrices associated with the cubic point group, it suffices to implement this addition to W for the e_g components only. The numerical results presented in this paper were obtained with this two-parameter approximation. The spin-polarized band structure used in the calculations is the same as that given in Ref. 1, where the two potential parameters were chosen to reproduce the measured moment and its symmetry character. With the band structure generated in this way, the cross section [Eq. (2)] is determined uniquely; i.e., there are no adjustable parameters. The cross-section expression was numerically evaluated with wave functions and electronic energies obtained from a self-consistent solution of the band-structure equation, and Brillouin-zone sums were evaluated by use of the tetrahedron method.

Results for **q** along [100] are shown in Figs. 1 and 2 in the form of the spin-wave dispersion curve and a contour plot of the scattering intensity. These results were obtained from constant-**q** calculations of the cross section as a function of energy. Peaks in the scattering cross section correspond to spin waves only if the energy of the peak coincides with a resonance in the particle-hole Green's function, i.e., to a particle-hole bound state. The most remarkable feature of these results is the prediction of "optic" modes with the energy of the upper branch reaching about 750 meV at the zone boundary. Notice also the relatively complicated nature of the contour plot at about half-

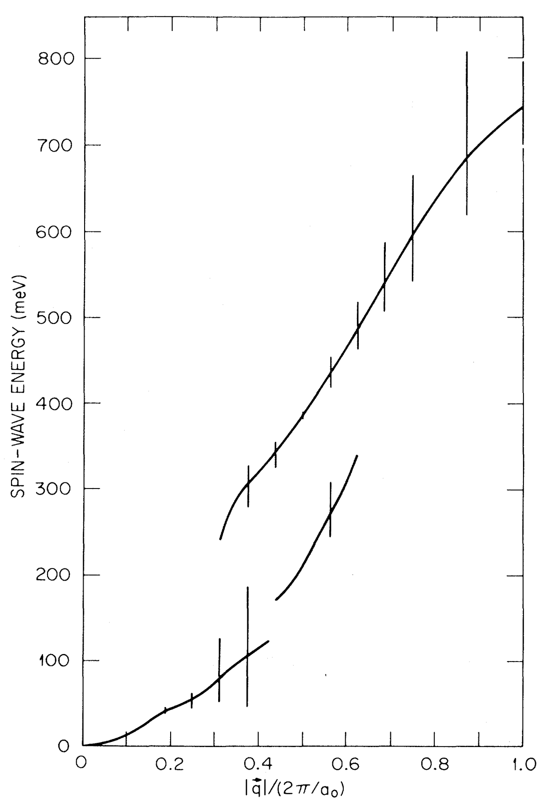

FIG. 1. Spin-wave dispersion curve for **q** along the [100] direction.

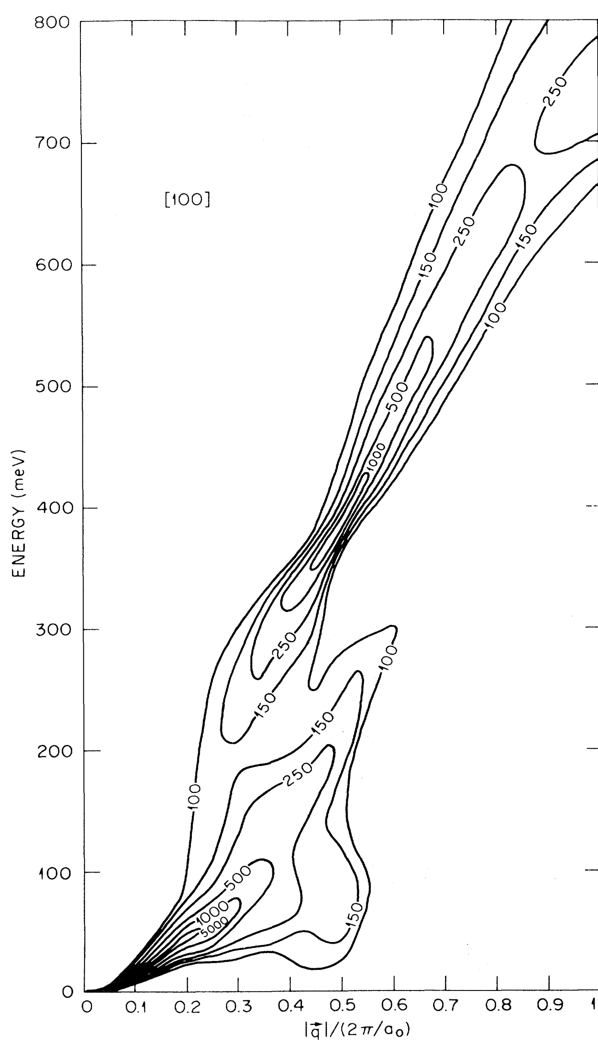

FIG. 2. Intensity contours, in arbitrary units, for **q** along the [100] direction.

way to the zone boundary. A plot of intensity against energy at $\mathbf{q}=(0.5,0,0)$ (units of $2\pi/a_0$) reveals a three-peak structure: a strong upper one at ~ 400 meV and two much weaker and broader ones at lower energies. The contour plot emphasizes the fact that the upper mode is quite sharp for both constant-$\mathbf{q}$ and constant-E scans. It is possible to express Eq. (2) as the sum of contributions with different symmetries. When this is done for $\mathbf{q}=(0.5,0,0)$, three of the terms in the sum show well-defined peaks, each one corresponding to a vanishing of the real part of a pole in the electron-hole Green's function. One can, therefore, understand the behavior in terms of three interesting spin-wave modes: two optic and one acoustic. The lowest-energy branch in Fig. 1 was not continued out to $|\mathbf{q}|=0.5$ because its corresponding peak is too broad to be considered a collective excitation. Because of problems associated with defining what is and what is not a spin wave, we feel that contour plots of the type given in Figs. 2 and 3 are a better way to display our results than the plotting of dispersion curves. With regard to the symmetry, the acoustic mode has mixed e_g and t_{2g} character, while the optic modes derive from interband transitions having virtually pure e_g character.

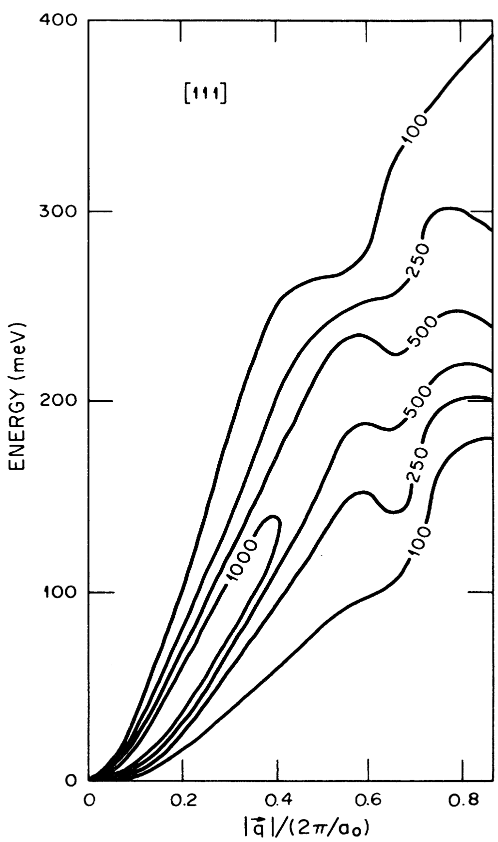

FIG. 3. Intensity contours, in arbitrary units, for $\mathbf{q}$ along the [111] direction.

Results for $\mathbf{q}$ along [111] are shown in Fig. 3 in the form of a contour plot of the scattering intensity. As in nickel[2] a single branch extends to the zone boundary. Energy transfers occur up to a little over 200 meV. Experiments[5] have been carried out with use of the spallation neutron source at Argonne National Laboratory to energies of 160 meV. Current time-of-flight measurements using spallation sources present what is essentially a composite of scatterings with a specific magnitude of q but for all directions. Results from the intense-pulsed-neutron-source experiment[5] could hardly be interpreted in terms of the [100] results shown in Figs. 1 and 2 but are in good agreement with results for $\mathbf{q}$ along [111] (Fig. 3), which apparently are dominating the experimental observation.

The qualitative form of the results given in Figs. 1 and 2 are fairly insensitive to the precise values of the parameters of the problem. Under small changes in the parameters, the three-peak structure at $\mathbf{q}=(0.5,0,0)$ remains, but the two weak components can become more or less well-defined and can vary considerably in their positions. The strong feature at $E\sim 400$ meV changes very little, however. The results for the [111] direction are more sensitive to small variations in the parameters. A two-branch structure, for example, is easily obtained in this direction. The single branch observed in the intense-pulsed-neutron-source experiment, however, appears to be consistent with the choice of W and the resulting band structure given in Ref. 1.

With regard to the sensitivity of the results to the form of the potential, we have also considered the effect of including $W_{\mu\nu\nu\mu}$ terms for the t_{2g} component. Preliminary results indicate that this can have fairly strong effects for the [111] and [110] directions but has no effect at all in the [100] direction. Calculations for more general forms of W are currently underway.

The experimental observation of the high-energy excitations is, therefore, likely to provide a very exacting test of models for both the band structure and the screened electron-electron interaction in the ferromagnetic transition metals. The number of modes found, along with their measured dispersion curves, will provide important information about the magnitude of the various W parameters. In addition, the search for the high-energy collective ("optic") excitations predicted in iron should provide an exciting challenge for experimentalists working at the limits of capability of reactor sources, and provide a novel high-energy-transfer experiment on a spallation source.

This research was sponsored in part by the Division of Materials Sciences, U.S. Department of Energy, under Contract No. DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc. One of us (J.F.C.) was on foreign assignment at the Rutherford Appleton

Laboratory, Chilton, Didcot, England. Another of us (T.M.) acknowledges encouragement of S. W. Lovesay and continuing support from the Rutherford-Appleton Laboratory.

[1]J. F. Cooke, J. W. Lynn, and H. L. Davis, Phys. Rev. B **21**, 4118 (1980).

[2]J. F. Cooke, J. A. Blackman, and T. Morgan, Phys. Rev. Lett. **54**, 718 (1985).

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

[4]H. A. Mook and D. Mck. Paul, Phys. Rev. Lett. **54**, 227 (1985).

[5]C.-K. Loong, J. M. Carpenter, J. W. Lynn, R. A. Robinson, and H. A. Mook, J. Appl. Phys. **55**, 1895 (1984).