Solid State Communications, Vol. 20, pp. 799–801, 1976. Pergamon Press. Printed in Great Britain

NUMERICAL INVESTIGATION OF SPIN WAVES IN FERROMAGNETIC IRON*

J.F. Cooke, J.W. Lynn† and H.L. Davis

Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830, U.S.A.

(*Received* 27 *May* 1976 *by H. Suhl*)

Spin wave energies and intensities have been calculated along three principal symmetry directions for ferromagnetic iron. These calculations are based on an itinerant model which incorporates band and wave-vector dependence of the relevant Coulomb matrix elements. The results indicate that iron's spin waves can be described completely by an itinerant model without recourse to additional assumptions about strong Hund's rule coupling or local moment behavior.

DURING the past few years neutron scattering experiments have yielded considerable information about spin waves in both ferromagnetic nickel and iron.[1–3] It has been known for some time that the itinerant-electron model of magnetism is capable of explaining, at least qualitatively, the unusual behavior exhibited by spin waves in these materials. However, a remaining important question has been whether the model would withstand fully quantitative tests. In this regard, a recent calculation of the generalized susceptibility showed that the itinerant model is capable of providing quite good quantitative agreement with low temperature spin wave data obtained from neutron scattering experiments for nickel.[4] In iron, however, the possible occurrence of strong Hund's rule coupling and/or local moment behavior might be expected to invalidate the applicability of the itinerant model. The purpose of this paper is to present some results which indicate that the itinerant electron model is also capable of providing a good quantitative description of iron's spin waves.

The present work is based on a slight generalization of the approximate (low temperature) expression for the transverse dynamic susceptibility, $\chi_T(\mathbf{q}, \omega)$, used in a previous nickel investigation.[4,5] This generalization leads to the following expression for the imaginary part of $\chi_T(\mathbf{q}, \omega)$:

$$\operatorname{Im} \chi_T(\mathbf{q}, \omega) = \text{const} \times \operatorname{Im} \sum_{\mu\nu=1}^{5} [I + \Lambda(\mathbf{q}, \omega - i\epsilon)]^{-1}_{\mu\nu} / U_\nu^{d-d}, \tag{1}$$

where

$$\Lambda_{\mu\nu}(\mathbf{q}, \omega) = U_\nu^{d-d} \times \sum_{\substack{nm\\ \mathbf{k}}} \frac{a_{n\mu\downarrow}(\mathbf{k}) a_{m\mu\uparrow}(\mathbf{k}+\mathbf{q}) a_{n\nu\downarrow}(\mathbf{k}) a_{m\nu\uparrow}(\mathbf{k}+\mathbf{q}) \{f_{n\mathbf{k}\downarrow} - f_{m\mathbf{k}+\mathbf{q}\uparrow}\}}{\hbar z - E(m\mathbf{k}+\mathbf{q}\uparrow) + E(n\mathbf{k}\downarrow)} \tag{2}$$

$$\begin{aligned} U_\nu^{d-d} &= U_{t_{2g}}^{d-d} \quad \nu = 1, 2, 3 \\ &= U_{e_g}^{d-d} \quad \nu = 4, 5. \end{aligned} \tag{3}$$

$E(n\mathbf{k}\sigma)$ is the electronic energy for band n, wave-vector $\mathbf{k}$, and spin σ, $f_{n\mathbf{k}\sigma}$ is the Fermi occupation number, and the $\{a_{n\mu\sigma}(\mathbf{k})\}$ are expansion coefficients of the Bloch functions in terms of symmetry orbitals. The $\{a_{n\mu\sigma}(\mathbf{k})\}$ and the electronic energy can be obtained from a solution of the energy band equations generated by the theory. We use $\mu = 1, 2, 3$ and $\mu = 4, 5$ for t_{2g} and e_g symmetry terms respectively. The energy band equations can be formally solved to give

$$E(n\mathbf{k}\sigma) = \sum_{\mu\nu} a_{n\mu\sigma}(\mathbf{k}) [\mathcal{H}_0(\mathbf{k})]_{\mu\nu} a_{n\nu\sigma}(\mathbf{k}) - \tfrac{1}{2} \times \sum_{\mu=1}^{5} U_\mu^{d-d} \{|a_{n\mu\sigma}(\mathbf{k})|^2 F_{\mu,\sigma} - |a_{n\mu-\sigma}(\mathbf{k})|^2 F_{\mu,-\sigma}\} \tag{4}$$

where $[\mathcal{H}_0(\mathbf{k})]_{\mu\nu}$ is the matrix element of an effective single particle Hamiltonian calculated with respect to the symmetry orbitals and $F_{\mu,\sigma}$ represents the number of electrons with symmetry character μ and spin σ. Equation (4) has been written in a form which demonstrates clearly the possible use of model Hamiltonian interpolation schemes to describe the band structure. An important feature of the generalization given by equations (1)–(4) is that the spin wave energy goes to zero as $\mathbf{q} \to 0$ regardless of the particular form of $[\mathcal{H}_0(\mathbf{k})]_{\mu\nu}$.[6]

There are two differences between the results given in equations (1)–(4) and those used previously.[5] First the spin dependence has been retained in the $\{a_{n\mu\sigma}(\mathbf{k})\}$

---

* Research sponsored by the U.S. Energy Research and Development Administration under contract with Union Carbide Corporation.

† Present address, Department of Physics, Brookhaven National Laboratory, Upton, NY 11973, U.S.A.

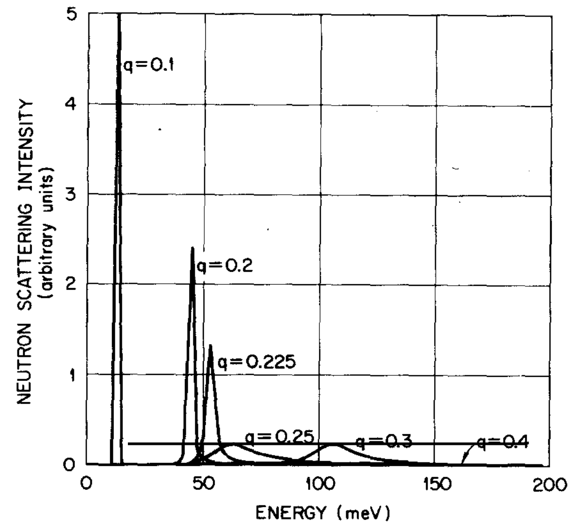

Fig. 1. Neutron scattering intensity for iron with **q** along the [100] direction. The wave vector $q = |\mathbf{q}|$ is measured in units of $2\pi/a$.

and second, we have allowed for the possibility that U^{d-d}, which can be related to the matrix element of a screened coulomb matrix element calculated with respect to the symmetry orbitals, might be different for e_g and t_{2g} symmetries.* Both of these generalizations follow directly from the theory. All of the information needed to calculate $\chi_T(\mathbf{q}, \omega)$ can be obtained from a solution of the energy band equations. Therefore, $\chi_T(\mathbf{q}, \omega)$ is uniquely determined once the ferromagnetic band structure is determined.

The first step in numerically evaluating equation (1) was to generate a paramagnetic crystal potential from a $3d^8$ atomic configuration. A second neighbor s–p–d Slater–Koster interpolation scheme was employed to generate the paramagnetic bands at arbitrary points in the Brillouin zone after the Slater–Koster parameters were determined from a least squares fit to a KKR first principles calculation. This procedure provides a fast and reasonably accurate method for generating the $\{[\mathcal{H}_0(\mathbf{k})]_{\mu\nu}\}$ matrix which appears in equation (4).

The ferromagnetic bands were obtained by solving the self-consistent equation generated by the theory.[5] The two parameters corresponding to U^{d-d} for e_g and t_{2g} symmetries respectively were chosen to produce the experimentally determined moment and ratio of e_g to t_{2g} character in the moment. The band structure which results from the procedure can be thought of in terms of a rigid splitting of 1.94 eV for pure d-symmetry states and zero splitting of pure s-like states. Even though the pure d-states are rigidly spin-split the $\{a_{n\mu\sigma}(\mathbf{k})\}$ turn out to be strongly **k**-dependent, and as a result of s–d hybridization effects the spin-splitting of the actual bands does exhibit a rather strong wave-vector dependence.

* Numerical estimates based on atomic-like orbitals indicate that the off-diagonal matrix elements between t_{2g} and e_g are small, and their effects have been neglected.

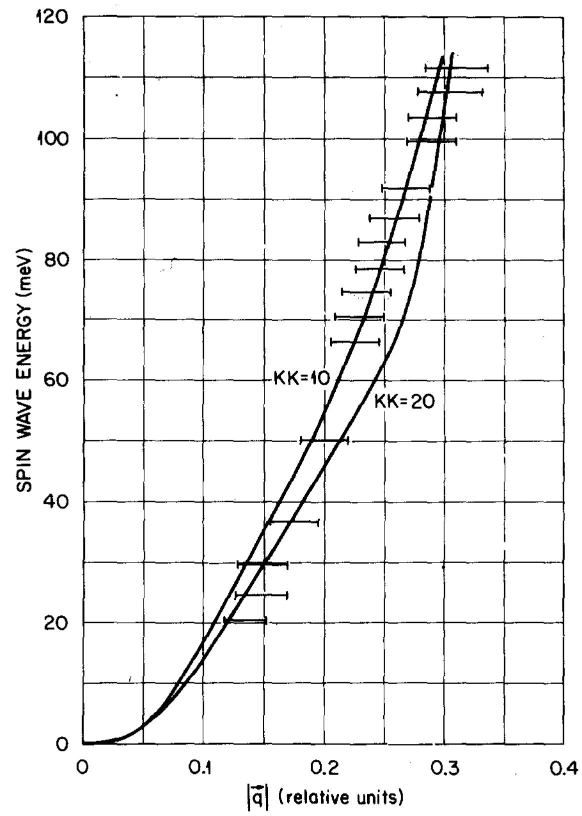

Fig. 2. Spin wave dispersion curve for iron. The wave vector, **q**, is measured in units of $2\pi/a$. Solid curves labeled KK = 10 and KK = 20 were obtained using a GR integration mesh of 440 and 3080 cubes respectively in the irreducible zone. The bars represent neutron scattering results of Mook and Nicklow.[2]

The numerical procedure for evaluating integrals of the type shown in equation (2) is based on the Gilat–Raubenheimer (GR) linear integration scheme and is the same as that used previously.[4] Six up-spin and six down-spin bands were used in the calculation. Because the Fermi energy falls in the middle of the d-bands the approximation of replacing $\{f_{n\mathbf{k}\downarrow} - f_{m\mathbf{k}+\mathbf{q}\uparrow}\}$ by one or zero in each of the small cubes used in the GR integration scheme generated some convergence problems in the calculation. In order to overcome these problems we found it necessary to use rather large numbers of cubes in the irreducible zone. We feel, therefore, that the numerical results which we have obtained do provide a reasonably good description of Im $\chi_T(\mathbf{q}, \omega)$.

Some of the zero temperature numerical results for the neutron scattering intensity [$\sim$ Im $\chi_T(\mathbf{q}, \omega)$] are shown in Fig. 1. We obtained similar results along both the [110] and [111] directions. The position of each peak determines the spin-wave energy for that **q**. As $|\mathbf{q}|$ increases the spin-wave peaks move to higher energy, broaden, and eventually disappear altogether for $|\mathbf{q}|$ somewhere between 0.35 and 0.4 (in units of $2\pi/a$). This broadening and ultimate disappearance of the spin-wave peak is due to the spin-wave running into a region where a particular weighted density of spin–flip excitations (Stoner excitations) is large.[6]

The spin-wave dispersion curve obtained from the spin-wave peak positions is shown in Fig. 2. The curves labeled KK = 10 and KK = 20 refer to the use of 440 and 3080 GR integration cubes respectively in the irreducible Brillouin zone. The bars represent the room temperature neutron data of Mook and Nicklow. The theory yields an isotropic spin wave dispersion curve, in agreement with experiment. This result is particularly noteworthy when considering the underlying band structure, since there is no obvious isotropy to the electron energy bands. The agreement between theory and experiment is not only qualitatively excellent, but quantitatively very good as well.

The intensity and width of the spin-wave scattering shown in Fig. 1 should also be directly comparable to neutron scattering experiments. Unfortunately, due to experimental difficulties there is no information of this type available at present for pure iron. Generally we can say, though, that the broadening of the spin waves above 50 meV may be large enough to be experimentally observable, and that the intensity of the scattering above ~ 120 meV will certainly be unobservable with the experimental sensitivity presently available.

The spin wave intensities have been measured for Fe (4 at.% Si) and Fe (12 at.% Si), whose magnetic properties are very similar to pure iron. Because of the steepness of the dispersion curves in these materials, the experimental data were obtained by fixing ω and varying **q**, rather than by fixing **q** and varying ω as in Fig. 1. Thus in order to directly compare with experiment we must first calculate the cross section at a large number of **q** points and then convolute this with the instrumental resolution, and the computer time required for this is prohibitive at present. Because the resolution employed in the experiments was by necessity rather coarse, we can nevertheless make a reliable estimate of the intensity by simply interpolating the cross section between different values of **q** and then evaluating the integrated intensity expected in the neutron measurements. The calculated intensities show a rapid fall off of the spin-wave scattering intensity at ~ 100 meV, which is in good agreement with experiment.

As a result of our calculations it appears that the itinerant model is certainly capable of providing good overall quantitative agreement with the neutron scattering results for iron at low temperatures. These results also indicate that the itinerant model is capable of describing systems with more than one unpaired-spin electron per site ($\mu_{\mathrm{Fe}} \sim 2.2\,\mu_B$) without recourse to additional assumptions about strong Hund's rule coupling. In order to extend the theory to finite temperatures one must go beyond the RPA Green's function decoupling scheme, which was used in the derivation of the result given in equation (1), and incorporate vertex corrections. It has been suggested that this approach might lead to the concept that the spin-splitting $[E(n\mathbf{k}\downarrow) - E(n\mathbf{k}\uparrow)]$ depends on a "local" magnetic moment which would exist in regions of short range order.[7] If this notion is correct it could account for the lack of temperature dependence displayed by the spin wave cut-off energy. Clearly more theoretical work is needed to resolve the finite temperature behavior of the itinerant model and more experimental work is needed to resolve the ambiguity in the spin-wave cut-off energy in iron.

REFERENCES

1. MOOK H.A., NICKLOW R.M., THOMPSON E.D. & WILKINSON M.K., *J. Appl. Phys.* **40**, 1450 (1969).
2. MOOK H.A. & NICKLOW R.M., *Phys. Rev.* **B7**, 336 (1973).
3. LYNN J.W., *Phys. Rev.* **B11**, 2624 (1975).
4. COOKE J.F. & DAVIS H.L., *AIP Conf. Proc.* **10**, 1218 (1973).
5. COOKE J.F., *Phys. Rev.* **B7**, 1108 (1973).
6. COOKE J.F. (to be published).
7. SOKOLOFF J.B., *Phys. Rev. Lett.* **31**, 1417 (1973).