CONF-801124--7(Draft)

NOTE: This is a draft of a paper being submitted for publication. Contents of this paper should not be quoted nor referred to without permission of the authors.

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering the article.

Temperature Dependent Dynamic Susceptibility Calculations

for Itinerant Ferromagnets

J. F. Cooke

MASTER

SOLID STATE DIVISION
OAK RIDGE NATIONAL LABORATORY
Operated by
UNION CARBIDE CORPORATION
Under
Contract No. W-7405-eng-26
With the
U. S. DEPARTMENT OF ENERGY
OAK RIDGE, TENNESSEE

October 1980

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

leg

Temperature Dependent Dynamic Susceptibility Calculations
for Itinerant Ferromagnets^a

J. F. Cooke

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

ABSTRACT

Inelastic neutron scattering experiments have revealed a variety of interesting and unusual phenomena associated with the spin dynamics of the 3-d transition metal ferromagnets nickel and iron. An extensive series of calculations based on the itinerant electron formalism has demonstrated that the itinerant model does provide an excellent quantitative as well as qualitative description of the measured spin dynamics of both nickel and iron at low temperatures. Recent angular photoemission experiments have indicated that there is a rather strong temperature dependence of the electronic spin-splitting which, from relatively crude arguments, appears to be inconsistent with neutron scattering results. In order to investigate this point and also the origin of spin-wave renormalization, a series of calculations of the dynamic susceptibility of nickel and iron has been undertaken. The results of these calculations indicate that a discrepancy exists between the interpretations of neutron and photoemission experimental results regarding the temperature dependence of the spin-splitting of the electronic energy bands.

PACS numbers: 71.25.Cx, 71.45.Gm, 75.30.Ds

INTRODUCTION

Inelastic neutron scattering experiments have revealed a considerable amount of information about the unusual spin dynamics of ferromagnetic nickel and iron (1-4). The low temperature spin dynamics obtained from these experiments have been shown to be in excellent quantitative as well as qualitative agreement with the itinerant model of magnetism (5). Theoretical predictions of the temperature-dependent spin dynamics of an itinerant model are very difficult to obtain and, until recently, were based on a simple temperature extrapolation of the low temperature theory (6). During the past few years, however, theoretical techniques have been developed which have yielded qualitative and, in some cases, quantitative agreement with the neutron scattering experiments at temperatures above the Curie temperature, T_C (7-10). The purpose of this paper is to present some results of calculations of the temperature dependent spin

dynamics of nickel and iron below T_c . In order to understand why such calculations are important, and what their results mean, we need to review some of the theoretical and experimental aspects of this problem.

In an itinerant ferromagnet the electrons which are responsible for the magnetic moment distribution occupy spin-split energy bands. The energy difference between opposite spin bands with the same wave-vector is called the spin-splitting energy, Δ . It has been suggested that as the temperature is increased Δ should decrease and vanish at T_c (6). In other words, the spin-split bands should collapse into a set of paramagnetic bands at T_c . This also implies that the magnetic moment should shrink to zero at T_c and, thus, there will be no moment (and no magnetism) for $T > T_c$.

The spin dynamics can be understood in terms of the two magnetic excitations which occur in an itinerant magnet. Stoner excitations are spin-flip excitations across the Fermi surface which create a hole of a given spin and an electron with opposite spin. These excitations occur primarily in a continuum of states called the Stoner continuum. The spin wave, on the other hand, is a bound state between an electron and hole with opposite spins. If the Stoner continuum lies low enough in energy then the spin wave will run into the Stoner continuum and disappear. If Δ decreases with temperature, then the Stoner continuum will be pulled down with increasing temperature and the energy at which the spin waves disappear should depend rather dramatically on temperature. The neutron experiments, however, show that this cut-off energy is essentially independent of temperature (1-4) and, therefore, Δ must have little or no temperature dependence.

This conclusion has been recently disputed by interpretations of angular resolved photoemission experiments (11-12). It has been inferred from these experiments that at the Curie temperature Δ has decreased by 40% in nickel and 20% in iron. In order to determine if a real inconsistency exists, it is clear that we need to determine the sensitivity of the spin-wave cut-off energy to changes in Δ and temperature.

This point has been investigated by performing temperature-dependent calculations of the dynamic susceptibility. The calculations which are reported in this paper were based on various extrapolations of the low temperature calculations of Cooke et al. (5). The results of these calculations provided an excellent description of the spin dynamics of both nickel and iron at low temperature and, therefore, serve as an excellent starting point for such

extrapolations. For purposes of clarity the remainder of this paper is divided into two sections. An outline of the general theory is given in the first section. The second section contains a summary of results and the general conclusions of the study.

GENERAL THEORY

Theoretical predictions of the spin dynamics can be obtained from the results of calculations of the dynamic susceptibility. The details of the low temperature calculations are published elsewhere (5) and will not be reproduced here. The calculations were based on a Green's function formalism and a generalized RPA decoupling scheme. The result obtained for the imaginary part of the transverse susceptibility, which is proportional to the inelastic neutron scattering cross-section, was found to be

$$\chi''(\vec{q}+\vec{\tau}, \omega) \sim \frac{d^2\sigma}{d\omega d\Omega} \sim \lim_{\epsilon \rightarrow 0} |F(\vec{q}+\vec{\tau})|^2 \times \sum_{\substack{\mu\nu \\ n}} (I + U\Gamma(\vec{q}, \omega + i\epsilon))_{\mu\nu}^{-1} \Gamma_{\nu n}(\vec{q}, \omega + i\epsilon) \quad (1)$$

where

$$(U\Gamma(\vec{q}, z))_{\mu\nu} = \frac{U_\nu}{N} \sum_{\substack{nm \\ \vec{k}}} \frac{a_{n\mu\uparrow}(\vec{k}) a_{m\mu\uparrow}(\vec{k}+\vec{q}) a_{n\nu\uparrow}(\vec{k}) a_{m\nu\uparrow}(\vec{k}+\vec{q})}{z - E(m\vec{k}+\vec{q}\uparrow) + E(n\vec{k}\uparrow)} \times \{f_{n\vec{k}\uparrow} - f_{n\vec{k}+\vec{q}\uparrow}\} \quad (2)$$

and $F(\vec{K})$ is a d-like form factor. In this expression $\vec{\tau}$ is a reciprocal lattice vector, and $\vec{q}$ is restricted to the first Brillouin zone. In the expression for the matrix $U\Gamma$, $E(n\vec{k}\uparrow)$ is the electronic energy for band n , wave vector $\vec{k}$, and spin $\uparrow$, $f_{n\vec{k}\uparrow}$ is the Fermi occupation number, and the $\{a_{n\mu\sigma}(\vec{k})\}$ are the expansion coefficients for an expansion of the electronic wavefunction in terms of symmetry orbitals

$$\psi_{n\vec{k}\sigma}(\vec{r}) = \sum_{\mu} a_{n\mu\sigma}(\vec{k}) \phi_{\mu}(\vec{r}). \quad (3)$$

The U_ν are constants which depend on the symmetry label ν . In order to calculate the susceptibility we must first determine the ferromagnetic band structure.

The electronic energy bands and expansion coefficients can be obtained by solving

$$\sum_{\nu} H_{\mu\nu}^{\sigma}(\vec{k}) a_{\nu\sigma}(\vec{k}) = E(n\vec{k}\sigma) a_{\mu\sigma}(\vec{k}) \quad (4)$$

where

$$H_{\mu\nu}^{\sigma}(\vec{k}) = H_{\mu\nu}(\vec{k}) \pm \Delta_{\mu} \delta_{\mu,\nu} \quad \begin{array}{l} + \text{ for spin } \uparrow \\ - \text{ for spin } \downarrow \end{array} \quad (5)$$

$H_{\mu\nu}(\vec{k})$ would be the appropriate Hamiltonian matrix if the system were not magnetic. The symmetry dependent constants Δ_{μ} are defined by

$$\begin{aligned} \Delta_{\mu} &= U_{\mu} (f_{\mu\downarrow} - f_{\mu\uparrow}) ; \quad d \text{ symmetry} \\ &= 0 ; \quad \text{otherwise} \end{aligned} \quad (6)$$

$$f_{\mu\sigma} = \frac{1}{N} \sum_{\vec{n}\vec{k}} f_{\vec{n}\vec{k}\sigma} |a_{\mu\sigma}(\vec{k})|^2$$

where $f_{\mu\sigma}$ is the number of electrons with symmetry μ and spin σ . The constants U_{μ} depend only on the irreducible representation to which μ belongs and, therefore, there are only two independent terms; one for e_g and the other for t_{2g} symmetry.

The band calculation now proceeds in two steps. The first step is to generate a "realistic" set of paramagnetic bands and use an interpolation procedure to determine the $H_{\mu\nu}(\vec{k})$. The second step is to determine the two constants for e_g and t_{2g} symmetries. These are chosen by requiring that the band calculation generate the correct spin-only moment and the correct symmetry character for the moment. Both of these static properties can be obtained from experiment (5). It is clear from Eqs. (1) and (4) that once the U_{μ} are determined, the band structure and the spin dynamics are uniquely determined.

The temperature dependent calculations which have been carried out thus far are based on simple temperature extrapolations of Eqs. (1) and (4). There are only two temperature dependent terms in the low temperature theory. One is, of course, the explicit temperature dependence of the Fermi occupation numbers

$$f_{\vec{n}\vec{k}\sigma} = \frac{1}{e^{\beta(E(\vec{n}\vec{k}\sigma) - E_f)} + 1} ; \quad \beta = \frac{1}{k_B T} \quad (7)$$

The other is a weak temperature dependence which the theory suggests might be associated with the U_{ν} . This dependence would result from changes in the local

magnetic field around lattice sites, which should occur as the temperature is increased. Some recent temperature dependent form factor measurements (13) indicate that these constants do indeed have a weak temperature dependence, which is different for the e_g and t_{2g} symmetry terms. These results have not yet been incorporated into the calculations. As in the zero temperature case, however, once the $U_d(T)$ and the temperature are given the band structure and the spin dynamics follow uniquely.

NUMERICAL RESULTS AND CONCLUSIONS

The numerical evaluation of Eq. (1) using the full ferromagnetic band structure is a difficult but straightforward computer calculation. The Brillouin zone sums defined in Eqs. (1) and (6) were carried out using the tetrahedron Brillouin zone integration scheme, with 2048 tetrahedra in the irreducible zone.

The low temperature ferromagnetic band structure for nickel has been generated from a set of paramagnetic bands proposed by Stocks et al. (14). The values of the parameters and a plot of the band structure along a few symmetry directions are given in Ref. 5. The band structure generated in this way has several interesting features. The energy band splitting was found to be strongly wave-vector dependent and, as a consequence of this only one d-like hole pocket was found at X in agreement with Haas-van Alphen experiments. In addition, the spin-splitting energy at L was calculated to be 0.3 eV which is in excellent agreement with the low temperature splitting obtained from recent angular resolved photoemission experiments (11,12). It should be pointed out that there was no fitting involved here since this ferromagnetic band structure was first published (15) before the experiments were performed. It is also important to note here that essentially all of the other band structures proposed for nickel have two hole pockets at X and twice as large a splitting at L.

The corresponding band structure proposed for iron was generated from paramagnetic bands proposed by Wood (16). In contrast to nickel, these bands were found to have approximately a rigid splitting of ~ 2 eV for the d-like states. This is somewhat higher than the 1.5 to 1.8 eV obtained from photoemission experiments but is still in reasonable agreement. As found in the case of nickel, the s-p-d hybridization effects generate a rather strong wave-vector dependent spin-splitting of the energy bands.

A number of different extrapolations of the low temperature band structure, and hence of the susceptibility, have been carried out. The simplest

extrapolation is, of course, to include only the explicit temperature dependence of the Fermi factors. The results of such calculations show that Fermi surface smearing has essentially no effect on the static or dynamic properties for iron. This follows from the fact that the Fermi energy lies in a fairly flat region of the density of states.

The case for nickel is quite different. The Fermi energy is in a very steep region of the density of states and, consequently, Fermi surface smearing should be more important. The calculations yielded two interesting and unexpected results. First, the low temperature expression for the magnetization

$$M(T) = \frac{S(T)}{S(0)} ; S(T) = \frac{1}{N} \sum_{\vec{k}} (f_{\vec{k}\uparrow} - f_{\vec{k}\downarrow}) \quad (8)$$

yields a Curie temperature of $1.7 T_C$, where T_C is the experimental result. Interestingly enough, if we call this an effective Curie temperature, T_C^* , and plot M as a function of T/T_C^* then the experimental magnetization curve is reproduced. Furthermore, it was found that $M(T_C) = 0.91$, which indicates that at T_C the average moment around each lattice site has decreased by about 9% as the result of Fermi surface smearing alone.

The second, and most interesting effect generated by the temperature dependence of the Fermi factors is related to the spin-wave dispersion curve. A calculation of the susceptibility along [111] reveals that Fermi surface smearing accounts for essentially all of the spin-wave renormalization up to $T_C/2$. For $T > T_C/2$ this effect becomes less important and other temperature factors must be included. The spin-wave cut-off energy, on the other hand, does not change with temperature, which is consistent with the neutron scattering results.

In order to investigate the compatibility of the conclusions drawn from neutron scattering and photoemission experiments concerning the temperature dependence of the spin-splitting energy, calculations of the susceptibility have been carried out which include the Fermi factor temperature dependence and the band splitting inferred from the photoemission experiments. The calculations were carried out at $0.8 T_C$ with $\vec{q}$ along [111], and the results are shown in Fig. 1. The closed and open circles refer to the neutron scattering data for 4.2°K and $0.8 T_C$ respectively. The solid line is the theoretical calculation for 0°K, and the dashed line corresponds to the calculation for $0.8 T_C$. Not only are the $0.8 T_C$ results in very poor agreement with the neutron experiments but the spin-wave cut-off

energy has been reduced to a value below 30 meV. It should be noted that these results are due entirely to the large reduction in the spin-splitting energy and not to the Fermi surface smearing.

A number of other calculations have also been carried out for various temperatures and splitting energies. By way of summary, the results of these calculations indicate that small changes in $U_d(T)$, coupled with the Fermi factor temperature dependence, can explain the entire spin-wave renormalization in nickel up to $2 T_c$. For iron, relatively large, and hence unrealistic, changes in $U_d(T)$ are required to generate the experimental dispersion curves.

There are several general conclusions which can be drawn from this study. The most important is that the spin-splitting energy can not be a strong function of temperature, so that the temperature dependence of Δ inferred from the angular resolved photoemission experiments is probably incorrect. It was also found that Fermi surface smearing accounted for essentially all of the spin-wave renormalization for $T < T_c/2$ in nickel and was unimportant in iron. Finally, in order to account for the spin-wave renormalization in iron (and nickel for $T > T_c/2$) additional temperature factors, such as spin wave - spin wave interactions and small changes in Δ with T , must be included into the calculation.

REFERENCES

a) Operated by Union Carbide Corporation under contract W-7405-eng-26 with the U. S. Department of Energy.

1. H. A. Mook, R. M. Nicklow, E. D. Thompson, and M. K. Wilkinson, J. Appl. Phys. 40, 1450 (1969).
2. H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. 30, 556 (1973).
3. H. A. Mook and R. M. Nicklow, Phys. Rev. B 1, 336 (1973); J. W. Lynn, *ibid.* 11, 2624 (1975).
4. J. W. Lynn, Ph.D. thesis, Georgia Institute of Technology, April 1974.
5. J. F. Cooke, J. W. Lynn, and H. L. Davis, Phys. Rev. B 21, 4118 (1980).
6. R. D. Lowde and C. G. Windsor, Adv. in Phys. 19, 813 (1970).
7. R. E. Prange and V. Korenman, Phys. Rev. B 14, 4691, 4698 (1979).
8. J. Hubbard, Phys. Rev. B 19, 2626 (1979).
9. H. Capellmann, Z. Physik B 34, 29 (1979).
10. M. V. You, V. Heine, A. J. Holden, and P. J. Lin-Chung, Phys. Rev. Lett. 44, 1282 (1980).
11. F. J. Himpsel, J. A. Knapp, and D. E. Eastman, Phys. Rev. B 19, 2919 (1979).

12. D. E. Eastman, J. F. Janak, A. R. Williams, R. V. Coleman, and G. Wendin, J. Appl. Phys. 50 7423 (1979).
13. J. W. Cable, private communication.
14. G. M. Stocks, R. W. Williams, and J. S. Faulkner, Phys. Rev. B 4, 4390 (1971).
15. J. F. Cooke and H. L. Davis, AIP Conf. Proc. 10 1218 (1975).
16. J. H. Wood, Phys. Rev. 126, 517 (1962).

FIGURE CAPTION

Fig. 1. Theoretical and experimental spin-wave dispersion curves for ferromagnetic Ni at $T = 0$ and $T = T_c/2$. The temperature calculations included only Fermi factor temperature dependence. a_0 is the lattice parameter.