MAGNETIC EXCITATIONS IN Fe AND Ni

To cite this version:

HAL Id: jpa-00228307
https://hal.science/jpa-00228307
Submitted on 1 Jan 1988

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
MAGNETIC EXCITATIONS IN Fe AND Ni

D. McK. Paul (1), H. A. Mook (2), P. W. Mitchell (3) and S. M. Hayden (4)

(1) Department of Physics, University of Warwick, Coventry CV4 7AL, G.B.
(2) Solid State Division, Oak Ridge National Laboratory, TN 37831, U.S.A.
(3) Department of Physics, University of Manchester, Manchester M13 9PL, G.B.
(4) Institut Laue-Langevin, 156X, 38042 Grenoble Cedex, France

Abstract. – Triple-axis neutron spectroscopy has been used to study the high energy magnetic excitations of iron and nickel. Excitations of energies up to 300 meV have been observed. The experimental results demonstrate clear evidence for interactions between spin-waves and single particle excitations as well as the existence of “multiple branches” of excitations.

The magnetic excitations of itinerant systems is a problem of considerable experimental and theoretical interest [1-5]. The important questions to be addressed relate to the energy range of the fundamental excitations of the spin system, how these excitations are modified by interaction with the single particle modes of the electronic spectrum and whether “state of the art” calculations based on band-structure models are capable of providing an adequate description of the experimental results. The experimental results described in this paper represent the most extensive study of the high energy magnetic excitations in itinerant systems which has been carried out to date and is restricted only by the limitations imposed by the neutron flux available at the currently available facilities.

Magnetic excitations in single crystal samples are measured using triple-axis spectrometers. This particularly flexible tool has been developed over the years and numerous examples of it’s use for the examination of the dispersion and lifetime of excitations in various materials are to be found in the literature. However for the study of excitations with energies greater than ~100 meV rather exacting design criterion are required to make the measurements feasible. To measure excitations up to some specific energy the incident energy of the neutron beam must be at least twice the excitation energy. This criterion is required to restrict spurious processes being detected but also to allow the measurements to be made at reasonable scattering angles. The reduction in intensity due to the decrease in form factor typically limits the experiment to working in the Brilouin Zone around the lowest order Bragg reflections. Therefore a large neutron flux with energies in the range 200-1000 meV is a necessity for these experiments. Additionally large monochromator and analyser crystals, preferably with the ability to focus the neutron beam on the sample, are required to make the best use of the neutron source. Various filters, thin plates of In, Hf, Cd, Sm etc, are required in the beam incident on the sample and in that scattered by the sample to restrict the observation of scattering processes involving the λ/2 contamination from the monochromator and also to absorb the low energy thermal neutrons which are present due to incoherent and inelastic scattering processes. The only machine which is capable of providing the required flux of high energy neutrons is the IN1 spectrometer of the Institut Laue-Langevin in Grenoble. The typical configuration used consists of a vertically focussing Cu monochromator set for reflection by the (331) planes and a vertically focussing Cu analyser crystal using the (220) reflection. Despite the suitability of this instrument for these experiments large single crystals are required to provide sufficient signal to observe the magnetic scattering to high energies. Isotope enriched samples were used to reduce the background due to incoherent and multiple phonon scattering. The crystals used in these experiments were 400 g of 60Ni and 200 g of 54Fe alloyed with 10 % of silicon to stabilise the bcc phase.

The spin wave dispersion relation was mapped to the highest accessible energy, or to the zone boundary if possible, for the [100] and the [111] directions in both materials [1, 2]. Some aspects of the experimental results are similar in the two materials. A contour plot of the magnetic scattering for iron in the [100] and [111] directions is presented as figure 1. The anisotropy of the dispersion relation is such that the excitations are at considerably higher energies in the [100] direction than in the [111] direction. In both materials the excitations near to the zone boundary in the [111] direction have considerable intrinsic widths (~20-40 % of the excitation energy) due to interactions with the Stoner or single particle modes. In the [100] direction an unusual structure in the dispersion relation is observed in both Ni and Fe, the spin wave branch is experimentally found to be divided into two portions. An extrapolation of the slope of the lower energy magnons gives a curve which describes the general trend of the spin wave dispersion but there exists a region of energy where either no spin waves exist.
or the spin waves are highly broadened. This unusual structure has led to discussions of the existence of an "optical" spin wave mode in iron and nickel, although such terminology may not be particularly illuminating. Theoretical calculations of the dynamical susceptibility based on accurate band structure models give a reasonable description of the general features found in the experiments, but are not yet able to exactly model the precise details [4, 5]. The calculations demonstrate the large anisotropy between the [100] and the [111] directions, estimate the magnon directions, estimate the magnon broadening and predict the "gap" in the [100] direction. The experimental data is currently being used to refine the modifications of the potentials used in the theoretical studies. These experiments directly demonstrate the influence of the electronic correlations on the propagation of spin waves in an itinerant system and stress that the effects produced are highly anisotropic thus demonstrating the dangers of interpretation which exist in examining polycrystalline materials with time-of-flight instruments. Such arguments also carry over to the observation of the magnetic fluctuations at high energies in the paramagnetic state.

A recent experiment [3], using the same single crystal sample of nickel, has attempted to investigate what happens to the high energy magnetic excitations as the temperature of the material is taken through the critical temperature for long range ordering. There exists considerable disagreement about what happens at low energies in such an experiment, with the emphasis being on whether spin waves develop from the diffusive scattering found near the zone centre. The resolution of the disagreement requires the discrimination of two similar lineshapes (one diffusive the other corresponding to severely broadened spin waves) and requires and experiment which is effectively beyond the current technology. In the present experiment we observed the changes in the excitation at [0.25, 0.25, 0.25] as a function of temperature. At room temperature, (0.46 $T_c$), this excitation is at 160 ± 15 meV with a width of 19 ± 15 meV, on increasing the temperature the excitation decreases in energy and the width increases, at 0.85 $T_c$ the excitation is measured to be at 135 ± 15 meV with a width of 27 ± 15 meV. Within the experimental accuracy and resolution no change in either the position or width of the excitation can be detected at 1.1 $T_c$, such a result is not really unexpected as $T_c$, on an energy scale, is only 55 meV but the well defined nature of the excitation suggests that we are observing a spin wave mode. This data maps well on to the previous examinations of the structure of the excitations in the paramagnetic state demonstrating the development of propagating excitations with widths much less than the excitation energy for energies greater than the energy equivalent of $T_c$. An obvious conclusion to be drawn from this experiment is that in the paramagnetic state itinerant systems still support excitations at high energies, just as they do at low temperatures. We would expect the excitations to be at even higher energies in the [100] and [110] directions and hence experiments which attempt to extract the degree of short range magnetic order above $T_c$, by measuring $\chi(q)$ require very large incident energies (at least 500 meV is required to integrate over the excitations in the [111] direction).

The experiments described in this paper represent the current "state of the art" in measuring excitations at high energies in itinerant systems, the limitations being imposed by the monochromator and analyser crystals currently available. Recent experiments on the magnetic excitations in cobalt on both the IN1 spectrometer and the HET time-of-flight spectrometer at the Rutherford Appleton Laboratory suggest that the study of single crystal specimens, with an accessible energy range matching and possibly exceeding that described here, may be possible using time-of-flight techniques [6].