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The magnetic response of paramagnetic Fe at high energy transfers

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Résumé. Des mesures antérieures dans le fer paramagnétique ont révélé une diffusion magnétique importante des neutrons à petits angles, la diffusion observée à grands angles étant très faible au contraire. Des mesures supplémentaires au voisinage du bord de la zone de Brillouin ont été effectuées jusqu'à des transferts d'énergie de 2 $T_c$. Une valeur de 1.7 $\mu_B$ est obtenue, conduisant à un moment intégré du fer de 1.55 $\mu_B$. Ces résultats confirment que dans les métaux 3d, des corrélations cohérentes au caractère itinérant des électrons 3d, sont présentes.

Abstract. Previous measurements in paramagnetic iron using polarized neutrons with polarization analysis revealed strong forward magnetic scattering. By contrast, the scattering at large wavevectors, was observed to be very weak. We report on additional measurements near the zone boundary, which extend to energy transfers of about 2 $T_c$ (~ 200 meV). A value of 1.7 $\mu_B$ is obtained, and the resulting integrated Fe moment is 1.55 $\mu_B$. These results confirm that, in 3d metals, correlations which are inherent to the itinerant nature of the 3d electrons occur.

1. Introduction.

Polarized neutron scattering with polarization analysis is a powerful tool for the study of the paramagnetic state in magnetic systems. In general, the neutron cross-section is proportional to the imaginary part of the generalized susceptibility $\chi''(Q, \omega)$ and when integrated over all energies, yields the isothermal susceptibility $\chi(Q)$.

In systems of local moments the bulk susceptibility $\chi(0)$ obeys a Curie-Weiss law,

$$\chi(0) = S(S + 1) \frac{T}{(T - \theta)},$$

where $\theta_C$ is the Curie paramagnetic temperature. While at large $Q$ where interference effects become negligible, $\chi(Q) = S(S + 1)$. Thus, in Heisenberg systems an enhancement of the magnetic scattering by the factor $T/(T - \theta)$ is observed in the forward direction, which reflects residual correlations in the paramagnetic state.

In 3d metals where the magnetism arises from itinerant electrons, the paramagnetic scattering of neutrons has revealed a behaviour which does not correspond to such a picture [1, 2]. Strong forward scattering has been observed in agreement with the large paramagnetic susceptibility of these systems, but the scattering at large $Q$, near the zone boundary, was found to be much lower than in Heisenberg systems. These results have been interpreted as due to ferromagnetic correlations arising from the delocalized nature of the electronic wavefunctions, and hence do not occur in systems of localized moments.

Recently, Wicksted et al. [3] reported measurements of the paramagnetic response at small $Q$ (i.e. $< 0.6 \text{Å}^{-1}$) in Fe. In this $Q$ range their measurements of the energy integrated intensities are in agreement with those obtained at the Institut Laue-Langevin. In preliminary measurements at large $Q$ they could not detect any significant magnetic signal, which underlines the weakness of the scattering in this $Q$-range. Strong low $Q$-fluctuations have also been observed in liquid Fe. A qualitative determination of the paramagnetic scattering by Nguyen et al. and Weber et al. [4, 5], confirmed the existence of a significant moment in the molten state. The scattering was found to be enhanced at small $Q$ showing that spin correlations remain relatively strong.
Strictly speaking, the deduction of \( x'(Q) \) from the neutron cross-section necessitates integration over energies up to infinity. In localized spin (Heisenberg) systems, it is fairly obvious that integration up to energies of the order of \( k_B T_e \) is sufficient, since nearly all the paramagnetic excitations are contained within this energy range. In the first measurements reported on Fe integration was only performed up to energies of about \( k_B T_e/2 \), supplemented by some constant \( Q \) scans extending to higher energies. It was argued by Edwards [6], that the difference between the observed behaviour and the predictions of a Heisenberg model had to be attributed to the truncation in the energy integration of the scattering. These arguments did not take into consideration the observed energy dependence of the scattering. In Edwards' model, the vast majority of the scattering falls within an energy range of \( \sim k_B T_e \) and thus predicts substantially more scattering in the energy range already covered. The discrepancy between the predictions of a Heisenberg model and the properties of itinerant systems had already been put forward by Herring [7] in his review article. In effect, in a Heisenberg system only thermal fluctuations are involved, since the single particle spectrum is well separated from the thermal energy region by several eV's. This is experimentally demonstrated by collective excitations of local moments or « magnons » which can propagate throughout the Brillouin zone in the ground state. In Fe the region of single particle excitations starts at zero energy and extends up to the bandwidth. Thus the paramagnetic response can be expected to extend to an energy much greater than \( k_B T_e \), particularly at large wavevectors since these correspond to local fluctuations.

As the most spectacular differences between Heisenberg and itinerant models are revealed at large energy transfers, we decided to perform some additional measurements of the paramagnetic scattering in Fe, in order to obtain a more accurate determination of the magnetic scattering, including energy transfers substantially higher than the range of thermal energies.

2. Experimental.

The sample used was the same single-crystal of Fe-5 at. % Si for which measurements have already been reported [1]. It is in the form of a cylinder of 9 mm diameter \( \times \) 50 mm long, with a \( \langle 110 \rangle \) direction parallel to the cylinder axis. It was placed in a furnace, heated by a molybdenum resistor of 38 mm dia, which fitted inside the Helmholtz coils used for controlling the polarization direction.

The intention of this experiment was to integrate over the thermal fluctuations, to obtain \( S(q) \) throughout the zone, making use of high incident polarized neutron energies > 0.5 eV (\( < 0.42 \text{ Å} \)) available on the D5 spectrometer. The intrinsically poor instrumental resolution obtained with short wavelengths has been intentionally compounded by the use of monochromating and analysing crystals having a large \( d \) spacing \( d_{111} (\text{Cu}_3\text{MnAl}) = 3.45 \text{ Å} \). Both monochromator and analyser were magnetized in the vertical direction. Guide fields maintained the neutron polarization between monochromator and sample and between sample and analyser. The Helmholtz coils around the sample position could be energized so as to rotate the neutron polarization at the sample either parallel or perpendicular to the scattering vector. The vertical and horizontal field required to rotate the neutron spins and offset the magnetic effect of the furnace was some 100 Oe and had a negligible effect on the sample. An r.f. coil in the incoming beam enabled the incident neutron polarization to be reversed. The collimations used in the measurements were 60'-40'-40'-60', corresponding to a slightly improved \( Q \) and energy resolution at the elastic position (\( \lambda = 0.84 \text{ Å} \)) as compared with previous experiments.

3. Measurements of the paramagnetic scattering by polarization analysis.

Polarized neutron scattering with polarization analysis enables an unambiguous separation of the magnetic component from other sources of scattering. This is because magnetic scattering may involve spin-flip processes and only the magnetic part of the scattering is sensitive to the angle between the direction of the neutron polarization and the scattering vector. If the two possible neutron spin states, with respect to the vertical \( z \) axis, are \(+\) and \(-\), then the following amplitudes are obtained:

\[
U^{++} = b - pS_x + BI_z \\
U^{+-} = -p(S_x + is_y) + B(I_x + iI_y)
\]

where \( b \) is the nuclear coherent scattering amplitude, \( I \) the nuclear spin operator and \( S \) the projection of the spin operator perpendicular to the scattering vector \( Q \).

An experimental arrangement very commonly used in order to measure paramagnetic scattering is to align the neutron polarization along \( Q \). According to relations (1) and (2) all the magnetic scattering in this instance is spin-flip and proportional to \( \frac{3}{2} \langle S_x S_y \rangle \). However, a more reliable measurement may be obtained by combining cross-sections measured with \( z \) successively along \( Q \) and perpendicular to it. The differences between these measured cross-sections are proportional to \( \frac{1}{2} \langle S_x S_y \rangle \), and all other sources of scattering vanish in the subtraction. In addition, since no mechanical movement is involved in these measurements, the backgrounds are equal and therefore also subtract out of the difference cross-section. For neutrons polarized parallel or perpendicular to the scattering vector the magnetic response function \( S(Q, \omega) \) is related to the observed intensity by

\[
\frac{d^2\sigma^{++}}{dQ d\omega} - \frac{d^2\sigma^{+-}}{dQ d\omega} = \frac{K_T}{6 \tau_0 \nu_c^2} N_m S(Q, \omega)
\]
where $V$ is the volume of the sample, $V_c$ the volume of the unit cell, $N_m$ the number of magnetic atoms per unit cell, $r_o$ the classical electron radius, $\gamma$ the magnetic moment of the neutron and $K_i, K_f$ the incident and final wavevectors.

Corrections for both incomplete polarization of the beam and flipping efficiency were made following the procedure outlined by Ziebeck et al. [8].

4. Results.

The measurements were performed at 1273 K (1.25 $T_c$), the temperature at which most of the previous data were obtained. At $\lambda = 0.84$ Å and with the spectrometer set in the elastic configuration, the broad peak due to ferromagnetic correlations which characterize the scattering around the $\langle 110 \rangle$ position was observed to be consistent with the previous data.

Using a fixed final energy of 116 meV ($\lambda_f = 0.84$ Å) the energy width of the scattering was measured at several $Q$-values in the second Brillouin zone. In each constant $Q$ scan (Fig. 1) the magnetic scattering is centred at zero energy transfer. The essential contribution is restricted to energies below 50 meV, i.e. within the thermal energy range. The residual intensity observed at the highest energy in the scan at $[0.775, 0.775, 0]$ can be attributed to the broad energy resolution of the spectrometer inherent when using short wavelengths and Heusler crystals. Since the majority of the spin fluctuations are in the small $q$ and small $\omega$ region significant intensity can be observed at non zero energy transfers, for a nominally large $q$, owing to contributions picked up by the tail of the resolution extending to small $q$.

It is of special interest to compare the results with those expected from theory in order to obtain an absolute value for the scattering near the zone boundary, where only magnetic one-site pair correlation functions are expected to contribute. In particular, for a Heisenberg system, the enhancement of the bulk susceptibility by magnetic interactions varies as $T/(T - T_c)$. At 1.25 $T_c$ the bulk susceptibility in Fe corresponds to a value for $\langle S_i S_j \rangle$ of 53 $\mu_B^3$ leading to 10 $\mu_B^3$ for the one-site pair correlation function. By contrast, integration of the data at $Q[0, 0, -1.2]$ yields a value of 0.7 $\mu_B^3$ which, after correction for the form factor and Debye-Waller factor, corresponds to only 1.5 $\mu_B^3$.

A direct measurement of this magnetic scattering may be obtained by opening the energy window of the spectrometer. This can be done when working with Cu$_2$MnAl crystals by increasing the incident and final energies of the neutrons. At $\lambda = 0.42$ Å, in the elastic configuration, the calculated energy resolution reaches 200 meV FWHM. However, the misalignment of the magnetic field with respect to $Q$, which occurs for large energy transfers, limits the scattering which can be effectively measured at low $Q$. But, for the large values of $Q$ which were chosen ($Q = 0, 0, 1.4$ and $Q = 1.4, 1.4, 0$) this effect becomes negligible. The measured magnetic signal after correction for the form factor and temperature factor reaches 1.9(0.3) $\mu_B^3$ and 1.8(0.3) $\mu_B^3$ respectively. Taking together all the measurements therefore leads to a mean magnetic signal at the zone boundary of 1.7 $\mu_B^3$ integrated over an energy width which reaches to nearly twice the thermal energy range. This value is in agreement with 0.8 $\mu_B^3$ obtained in previous measurements working at $\lambda = 0.84$ Å with 50 meV FWHM. The increase in the measured intensity results from the larger energy width. It is worth noting that at the zone boundary the magnetic signal is considerably smaller than expected for Heisenberg systems. The integration of the

Fig. 1. — Constant $Q$-scans in Fe at $[-0.775, -0.775, 0]$ and $[0, 0, -1.2]$ using a final wavevector of 7.48 Å$^{-1}$ and energy resolution of 30 meV FWHM. The large energy width observed in the scan at $[-0.775, -0.775, 0]$ is a consequence of the poor $Q$ resolution as explained in the text.
signal out to the zone boundary yields a magnetic moment of 1.55 $\mu_B$. Half of this contribution is contained within the low-$Q$ region ($< 0.7 \text{ Å}^{-1}$) which represents the ferromagnetic correlations characteristic of 3d itinerant systems.

5. Discussion.

These measurements confirm results previously obtained in the paramagnetic state of iron. The observation of a sharp quasi-elastic forward peak and similar sharp peaks at the Bragg positions show that the magnetic carriers giving rise to paramagnetic scattering have ferromagnetic correlations over several interatomic distances, even well above $T_c$. From the width of the forward peak, the characteristic wavelength ($2\pi/q_c$) of the correlations is deduced to be about 16 Å. These short range correlations are characteristic of metallic 3d magnets and have also been observed in nickel and in several transition metal rich intermetallic compounds [2].

At large wavevectors the paramagnetic scattering in 3d metallic magnets is, unlike the scattering at small wavevectors, strongly reduced as compared to local moment systems in their paramagnetic state. This absence of a substantial low frequency component in the scattering at large wave vectors could be anticipated from consideration of the magnetic response; in the scattering at large wavevectors one is essentially looking at fluctuations within a single atom, which in itinerant systems can extend in energy up to the bandwidth. This can be contrasted with the local moment system described by a Heisenberg model in which the thermal and one electron fluctuations, i.e. excitations changing the amplitude of the local moments, are well separated in energy. There the paramagnetic scattering falls within a window of $\sim k_B T_c$. If a Heisenberg model is used to fit the observed spin wave dispersion in Fe below $T_c$ it will give rise to magnetic fluctuations, the energy spread of which will be contained within a range of order $k_B T_c$ at the zone boundary. However, the observed magnetic weight within that energy window is substantially less than that predicted by such a model.

The peculiar features of itinerant electron systems have been interpreted in two conflicting ways. Shastry et al. [10], Hubbard [11] and Hasegawa [12], starting from fairly disordered configurations of the magnetic moments, obtained equilibrium states of 3d metals above $T_c$ which show little short range magnetic order. Prange and Korenmann [13], Moriya [14] and Capellmann [15] consider the itinerant nature of the electrons as essential to obtain a good description of the paramagnetic state. The phase coherence of the electronic wave functions extending over distances considerably larger than the interatomic spacing implies that short range magnetic order is intrinsic to the paramagnetic state of itinerant systems. It is now clear that this description is more closely in agreement with the experimental observations obtained by neutron scattering.

In conclusion the paramagnetic scattering from an itinerant electron magnet may be characterized in terms of two domains of momentum transfer. At small $Q (< 0.9 \text{ Å}^{-1})$ i.e. at long wavelengths, the microscopic properties of the d electrons are not resolved and behave similarly to a localized system. At high $Q$, the concept of a local moment is not relevant as in an insulating material and any attempt to describe the behaviour of metallic 3d magnets must consider the itinerant nature of the magnetic electrons. At momentum transfers close to the zone boundary one is essentially looking at fluctuations within a single atom, which in itinerant systems can extend in energy up to the bandwidth. This can be contrasted with the local moment system described by a Heisenberg model in which the thermal and one electron fluctuations, i.e. excitations changing the amplitude of the local moments, are well separated in energy. There the paramagnetic scattering falls within a window of $\sim k_B T_c$. If a Heisenberg model is used to fit the observed spin wave dispersion in Fe below $T_c$ it will give rise to magnetic fluctuations, the energy spread of which will be contained within a range of order $k_B T_c$ at the zone boundary. However, the observed magnetic weight within that energy window is substantially less than that predicted by such a model.

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