Paramagnetic scattering from Fe(3.5 at. % Si): Neutron measurements up to the zone boundary

G. Shirane, P. Böni, and J. P. Wickstead*
Brookhaven National Laboratory, Upton, New York 11973
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Paramagnetic scattering cross sections of Fe(3.5 at. % Si) are measured at $T=1.1T_c$ and $1.25T_c$ over the entire Brillouin zone. The major part of these magnetic cross sections is confined below 60 meV even near the zone boundary, in agreement with the recent report by Brown *et al.* The energy-integrated value of the magnetic moment at the zone boundary is $(2.7\pm1.0)\mu_B^2$. This value is smaller than what would be expected for localized systems and gives experimental support for the itinerant-electron character of ferromagnetism in Fe.

I. INTRODUCTION

The paramagnetic scattering from $3d$ ferromagnets, in particular, iron, has been the subject of intensive neutron scattering studies in recent years.[1–4] The use of polarization analysis,[5] i.e., polarized neutron beams, has greatly improved the detectability of the magnetic cross sections even in the presence of much larger nuclear cross sections. Very recently Wicksted, Böni, and Shirane[4] have reported extensive measurements on pure Fe at $T=1.02T_c$ and $1.06T_c$ and their constant Q measurements covered a q range of 0.1–0.6 Å^{-1} [(110) zone boundary is 1.54 Å^{-1}] and an energy range extending up to 50 meV. This paper extends the q range to the entire Brillouin zone. A large, cylindrical (40 mm in length and 20 mm in diameter) Fe crystal containing 3.5 at. % of Si, was utilized for these measurements.

The main conclusions of Ref. 4 can be summarized as follows. The constant Q measurements follow, up to $\zeta=0.15$, closely, the predictions of a simple double-Lorentzian scattering function

$$S(q,\omega)=\frac{1}{6}M^2(Q)\frac{1}{\pi}\frac{\Gamma}{\Gamma^2+\omega^2}\frac{\omega/kT}{1-e^{-\omega/kT}}\ , \tag{1}$$

$$M^2(Q)=4S(S+1)\frac{\chi(0)}{\chi_1}\frac{\kappa_1^2}{\kappa_1^2+q^2}\ , \tag{2}$$

where κ_1 is the inverse correlation length and χ_1 is the susceptibility of a system of noninteracting spins. To visualize this result we have redrawn in Fig. 1 the calculated intensity contours from Ref. 4, which parametrize the data well, at least up to $\zeta\simeq0.15$. The magnetic scattering cross sections

$$\frac{d^2\sigma}{d\omega\, d\Omega}=\gamma_0^2 f^2(Q)e^{-2W}S(q,\omega) \tag{3}$$

can be put on an absolute scale using measured phonon intensities from the same sample.[4] $M^2(0)$, extrapolated from our neutron measurements, compares well with the dc susceptibility $\chi(0)$ at high temperatures

$$M^2(0)=12kT\chi(0)\ . \tag{4}$$

The most important and surprising result of Fig. 1 is the validity of the simple expressions, Eqs. (1) and (2), for a wide range of q up to $\zeta=0.15$. The contour lines which result, exhibit relatively sharp peaks for constant E scans; these peaks were originally interpreted to be the signature of propagating spin waves[6] above T_c. It is now clear that these peaks are nothing but simple energy slices of the paramagnetic scattering function.[4]

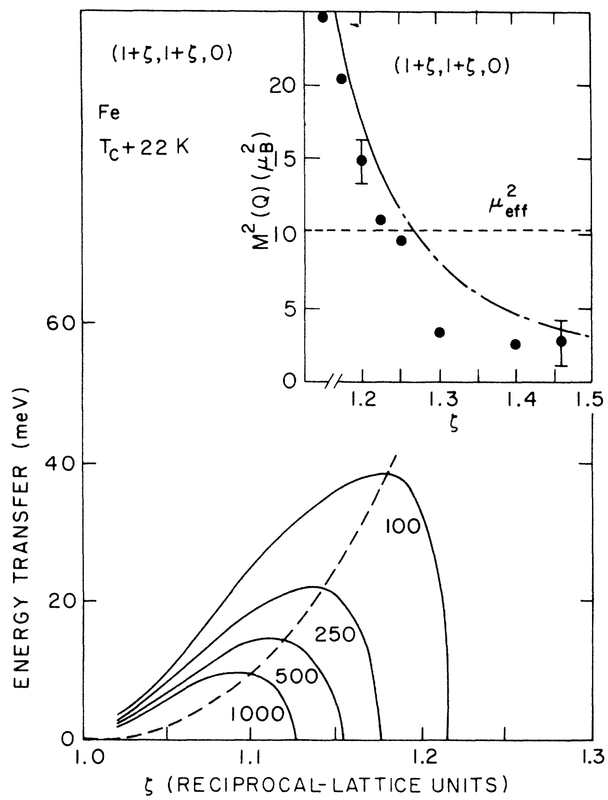

FIG. 1. Magnetic cross sections of pure Fe at $T=T_c+22$ K. The intensity contours in units of $10^{-3}\mu_B^2$/meV sr have been calculated using a modified double-Lorentzian scattering function, after Wicksted *et al.* (Ref. 4). The inset shows $M^2(Q)$ values extending up to the zone boundary (Ref. 4). The dashed-horizontal line represents ideal paramagnetic scattering. The dashed-dotted line corresponds to the q region where the Lorentzian behavior of $M^2(Q)$ is no longer expected to be valid.

One very important issue which was not settled by Wicksted *et al.*[4] was the proper line shape of the magnetic scattering near the zone boundary. Integrated intensities are given in the inset of Fig. 1, with rather large error bars. The moment near the zone boundary was estimated to be $(2.5\pm1)\mu_B^2$. This relatively small value was interpreted as a consequence of insufficient energy integration (up to 60 meV).

This same problem of the zone-boundary moment has been studied by Brown *et al.*[1,2] using a slightly different technique. They intentionally utilized neutron energies of 120 meV or larger, and, consequently, broader energy resolutions. Thus the polarization analyzer set at $\Delta E = 0$ integrated the magnetic cross sections within $2\Gamma_{res}$ (full width at half maximum) of the spectrometer energy resolution. With $2\Gamma_{res}=43$ meV, they reported[1] the zone-boundary value to be considerably lower than $1\mu_B^2$; a more recent study,[2] with $2\Gamma_{res}=200$ meV, quoted the value at $1.7\mu_B^2$. They also reported a few constant Q scans near the zone boundary and concluded that the significant part of the magnetic scattering is limited to the energy range below 60 meV. It is this question of the magnetic cross section near the zone boundary that we are addressing in this present paper.

II. POLARIZED-BEAM STUDIES

The details of the polarization analysis which result from our current setup were recently described,[4] and we emphasize only the essentials here. The magnetic cross sections are separated out from all other cross sections by simply switching the neutron polarization from a parallel direction (HF) to a perpendicular direction (VF), with respect to the scattering vector, and then taking the difference. The major difficulty in this triple-axis polarized-beam measurement is the drastic cut in intensities which results from the poor reflectivities of the Heusler polarizer and analyzer. This, however, is amply compensated in many experimental situations by the simplicity and beauty of this technique.

Figure 2 illustrates examples of relatively high-resolution scans taken with a final fixed energy $E_f=32$ meV. The phonons show sharp profiles even at this elevated temperature and can be utilized to put the paramagnetic cross sections onto an absolute scale. We discovered that the calibrations should be done by using phonons measured at the same temperature as the magnetic scattering. This is due to (1) the proper compensation of the Debye-Waller factor W in Eq. (3) and in the phonon cross sections, and (2) the effective-beam depletion, resulting from various scattering processes in the sample, which is strongly temperature dependent at high temperatures for large samples.

Figure 3 shows three selected profiles at intermediate q

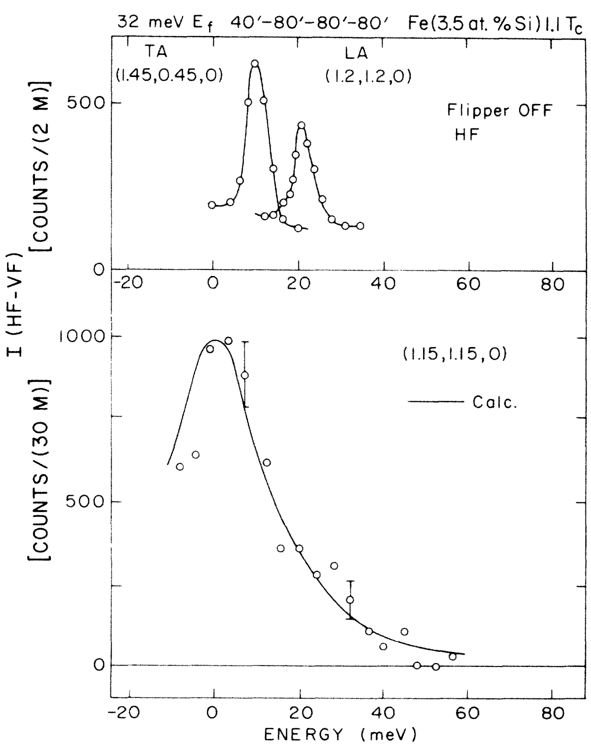

FIG. 2. Examples of high-resolution polarized-beam studies of Fe(3.5 at.% Si). Several phonons, as shown in the top part, are utilized to put the paramagnetic cross sections onto an absolute scale. M in ordinate label denotes the numerical value 10^6.

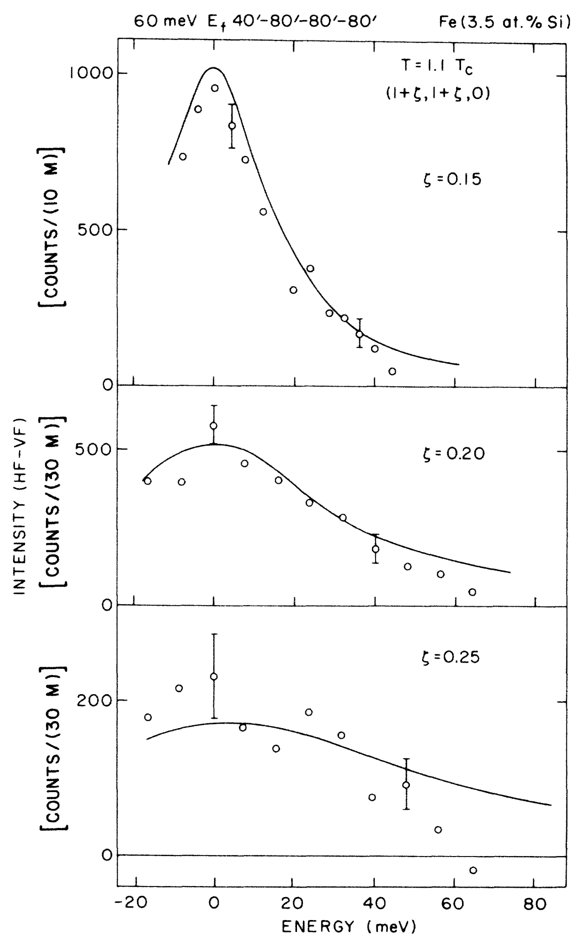

FIG. 3. Constant Q profiles at higher q's obtained with a more relaxed energy resolution; FWHM = 15 meV. 10 M monitor corresponds to 10 min at $\Delta E = 0$.

values obtained using a more relaxed resolution ($E_f = 60$ meV). The solid lines are calculated from the simple double Lorentzian given in Eqs. (1) and (2) with a *single* normalization factor fixed at $\zeta = 0.17$ (not shown). The agreement between this simple model and the observed cross sections is excellent up to $\zeta = 0.20$. Then we notice clear deviations starting at $\zeta = 0.25$. The linewidth of the data is narrower than the value predicted by the scaling law

$$\Gamma = Aq^{2.5} f(\kappa_1/q) , \tag{5}$$

where f is the dynamical scaling function given by Résibois and Piette.[7] This is expected since the q^2 expansion is limited to a small-q region and a natural change-over for the cross section, in its simplest form,[8] would be $1-\cos(2\pi\zeta)$. This was successfully demonstrated[8] for the localized ferromagnet Pd_2MnSn.

The measurements near the zone boundary suffer from severe intensity problems. We summarize all of the long counting scans (60 M monitor corresponds to 60 min at $\Delta E = 0$ and 120 min at $\Delta E = 60$ meV) in Fig. 4 for $T = 1.1T_c$ and $1.25T_c$, obtained at various E_f values between 42 and 60 meV. There are considerable fluctuations in the data points; nevertheless it is clear that the major cross sections lie below 60 meV. This is the most important conclusion of our current study and it is in full agreement with the data presented by Brown *et al.*[1,2]

Integrated intensities, converted into the moments $M^2(Q)$ in units of μ_B^2 [see Eqs. (1)–(3)], are shown in Fig. 5. The values $M^2(0)$ are extrapolated by Eq. (2), and are in good agreement with the values estimated from the dc susceptibilities shown by the shaded areas.[9] For iron with Si added, there are some ranges of $\chi(0)$ that one can estimate from the literature as listed in Table I. Solid and

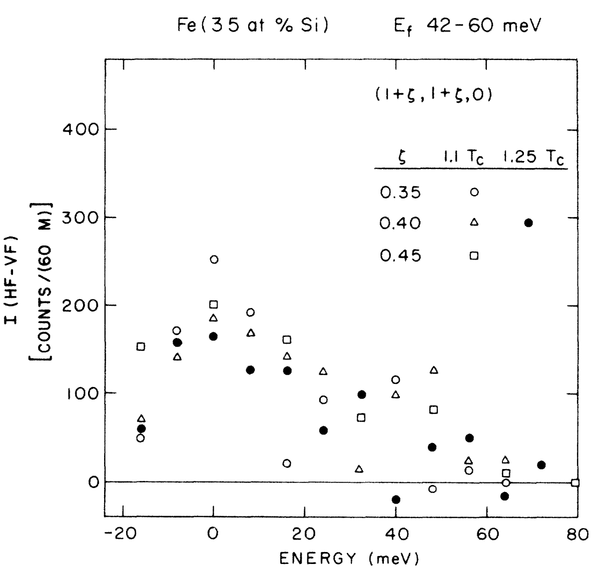

FIG. 4. Summary of constant Q profiles near the zone boundary taken under various conditions as listed in the figure. Major cross sections are confined below 60 meV. Typical error bars are ±50.

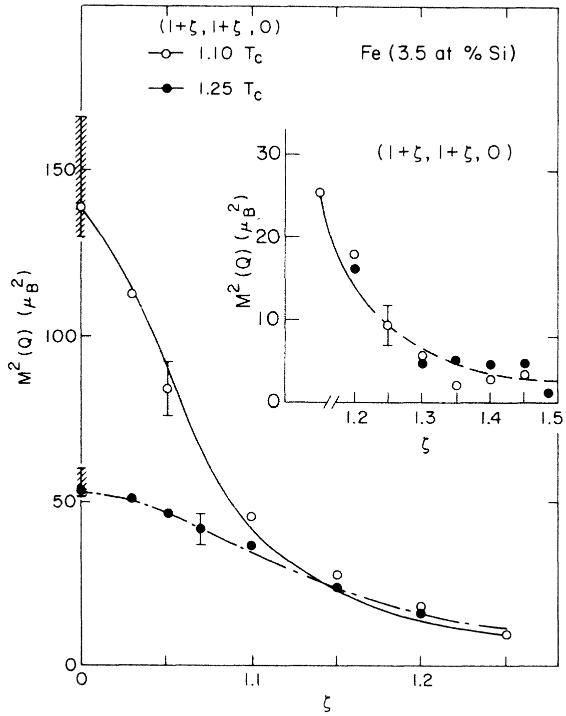

FIG. 5. Integrated intensities for the entire Brillouin zone. The values shown at $\zeta = 0$ are extrapolated values of our fits and they are in good agreement with the dc susceptibilities (see Table I). The inset shows $M^2(Q)$ values extending up to the zone boundary. The lines are explained in the text.

dashed-dotted lines shown in Fig. 5 are q^2 expansions [see Eq. (2)]. These, of course, are *not* expected to be valid at higher ζ's, and thus are shown as dashed lines in the inset. Our current results on Fe(3.5 at. % Si) are consistent with those for pure Fe shown in Fig. 1. The q-integrated value of $M^2(Q)$ over the Brillouin zone is about 70% of the value for $\mu_{\rm eff}^2$ ($\approx 9.73\mu_B^2$) obtained from the Curie constant. If the local-moment picture would prevail for Fe, the cross sections should be considerably higher than the dashed line, as demonstrated[8] for the Heusler alloy Pd_2MnSn.

The linewidths at $1.1T_c$ are summarized in Fig. 6. As previously stated, beyond $\zeta = 0.2$, they clearly deviate from the scaling law given by Eq. (5) and saturate around $\Gamma = 40$ meV. This result is quite unexpected from our previous study;[4] we assumed that Γ follows the lines A or B shown in Fig. 6. We carefully examined the experimental caveats which could cause the fall off of intensity at high-energy transfer. Under identical experimental condi-

TABLE I. Properties of Fe(3.5 at. % Si), $T_c = 1032$ K, $A = 142$ meV Å$^{2.5}$, $d^*(110) = 3.07$ Å^{-1} at T_c.

	$T = 1.1T_c$	$1.25T_c$
$M^2(0)$ from $\chi(0)$	$130-166\mu_B^2$	$53-58\mu_B^2$
κ_1	0.21 Å^{-1}	0.40 Å^{-1}

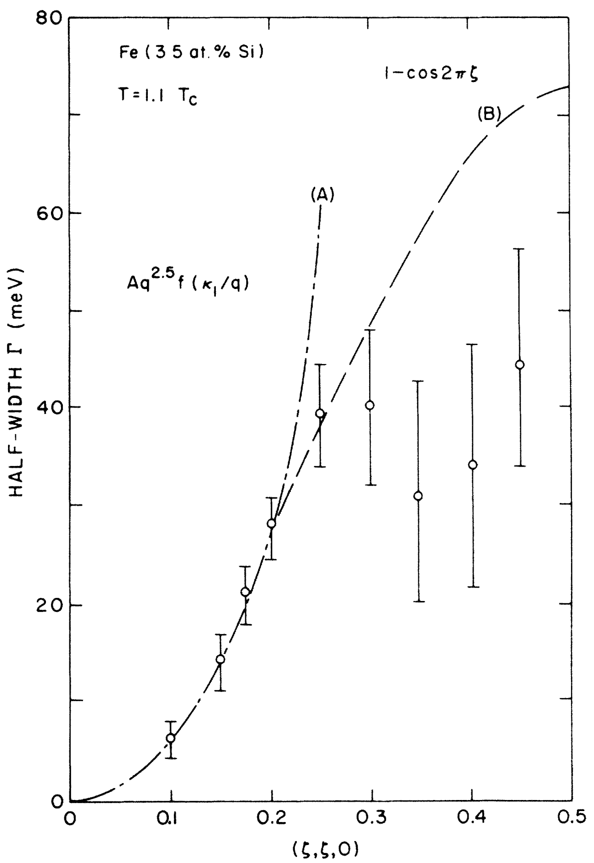

FIG. 6. Measured linewidths Γ compared with model calculations. The line *A* represents the dynamical scaling law and is valid up to $\zeta=0.2$. Line *B* is the $1-\cos(2\pi\zeta)$ description of the linewidth which gives a good characterization of the data for the localized ferromagnet Pd_2MnSn.

tions the measurements[10] on Ni show essentially flat cross sections up to 80 meV.

III. DISCUSSIONS

We have demonstrated the following for Fe(3.5 at.% Si):

(1) The simple double-Lorentzian cross sections [Eqs. (1) and (2)] give excellent representations for the magnetic cross sections up to $\zeta=0.20$.

(2) The linewidth Γ saturates at higher ζ's to a value of ≈40 meV.

(3) The major magnetic cross sections are confined below 60 meV, even at the zone boundary, and give a zone-boundary moment of $(2.7\pm1.0)\mu_B^2$, in full agreement with Brown *et al.*,[2] but considerably lower than the value of μ_{eff}^2, the value expected for an ideal paramagnet.

We have been looking for signatures of the itinerant nature of ferromagnetism in Fe from neutron scattering cross sections and item (3) is the first concrete experimental evidence that has been observed. We note that the above three items have also been seen to hold for selected constant-Q scans conducted along the [100] direction.[11]

Let us now compare our results with those reported by Brown *et al.* for the 5 at.% Si alloy, summarized in Fig. 7. Several parts of $M^2(Q)f^2(Q)$ are taken from the earlier publication obtained with $2\Gamma_{\text{res}}=43$ meV. The inset shows one example of a relatively narrow linewidth at high ζ. From the data in Fig. 7 near the zone boundary $(\frac{1}{2},\frac{1}{2},0)$, Brown *et al.*[1] first concluded an extremely small $M^2(Q)$. More recently, they[2] extended the measurements using a larger $2\Gamma_{\text{res}}$ of 200 meV and revised the value of the moment at the zone boundary to $1.7\mu_B^2$. This value is consistent with our value of $2.7\mu_B^2$ within both experimental errors.

Current theories[12] on magnetism in Fe do not give an explicit $S(Q,\omega)$, which we can compare directly with our results. It is not possible to explain the small cross sections near the zone boundary within the framework of the Heisenberg model. Thus we have to wait for quantitative theoretical calculations to establish an adequate model for magnetism in Fe.

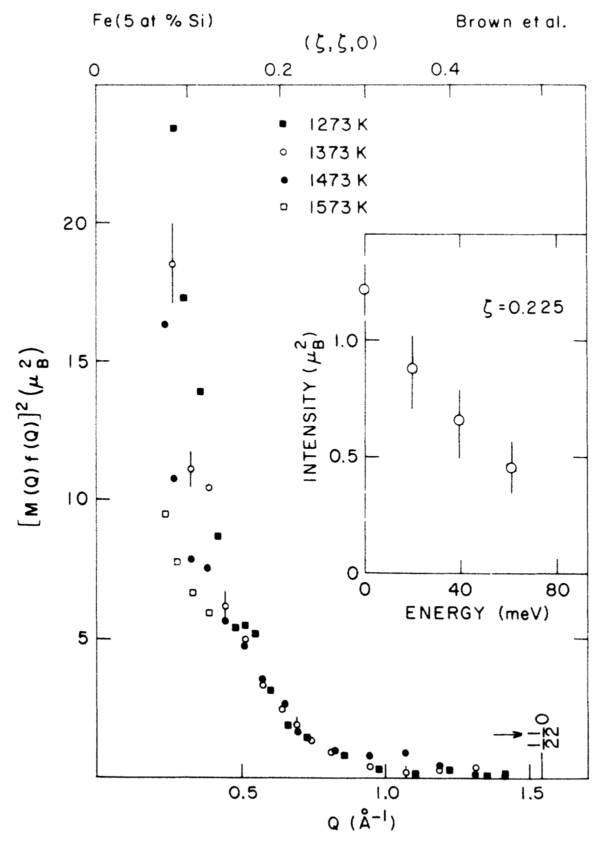

FIG. 7. Summary of magnetic cross sections reported by Brown *et al.* (Refs. 1 and 2) for Fe(5 at.% Si). The form factor $f(Q)$ must be removed for comparison with our Fig. 5 ($f^2=0.81$ at $\zeta=0.2$). The horizontal arrow at the zone boundary indicates the revised value of the moment (from Ref. 2). The inset shows a constant-Q scan at $\zeta=0.225$.

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*Present address: Department of Physics, Oklahoma State University, Stillwater, OK 74078.

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