Temperature-Induced Magnetism in FeSi

G. Shirane and J. E. Fischer[(a)]
Brookhaven National Laboratory, Upton, New York 11973

Y. Endoh
Tohoku University, Sendai 982, Japan

and

K. Tajima
Keio University, Yokohama 223, Japan
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We report neutron-scattering measurements with polarization analysis on a large single crystal of FeSi up to 650 K. A strongly q-dependent magnetic scattering is observed around (011) and the intensity follows the unusual shape of the static susceptibility. Surprisingly, the energy-integrated magnetic cross sections are almost q independent. This represents an entirely new kind of magnetism which, overall, supports the picture of temperature-induced moment in FeSi.

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Neutron scattering is a powerful probe of magnetic systems. For example, the usual signatures of ferromagnetism are magnetic Bragg peaks which reveal the long-range magnetic order, and structure in $S(q,\omega)$ from polarized-beam measurements which reflects the symmetry of magnetic correlations. FeSi and MnSi crystallize in a cubic structure (space group $P2_13$) in which magnetic atoms are located at displaced face-centered positions. MnSi is a spiral ferromagnet with the Curie temperature $T_C = 29$ K. This fascinating crystal has been extensively investigated by Ishikawa *et al.*[1] and it is now considered a prototype of a weak itinerant-electron ferromagnet.[2] FeSi, on the other hand, does not order magnetically. It shows, however, a very unique magnetic behavior which, until now, remains essentially unexplained.

In 1967 Jaccarino *et al.*[3] reexamined the anomalous properties of FeSi, in particular the dramatic temperature dependence of the susceptibility reproduced here at the top of Fig. 1. They proposed that FeSi is a nearly ferromagnetic small-gap semiconductor as an alternative to models involving strongly exchange-coupled isolated spins or an antiferromagnetic phase below a (rather high) ordering temperature. The gap model was supported by a band calculation by Nakanishi, Yanase, and Hasegawa,[4] while a spin-fluctuation model by Takahashi and Moriya[5] gave a satisfactory account for $\chi(T)$ and the specific heat. Evangelou and Edwards[6] then emphasized the itinerant-electron nature of FeSi and pointed out the likelihood of ferromagnetic correlations. Neutron-scattering experiments[7-9] prior to 1983 failed to detect any magnetic scattering. More recently Ziebeck *et al.*[10] observed a magnetic signal from FeSi powder using polarization analysis; their 500-K data are reproduced as the inset to Fig. 2. The increase in magnetization at small q was interpreted as a signature of ferromagnetic correlations, while it was claimed that the coupling changes to antiferromagnetic at low temperatures.

In this paper we report polarized-neutron results on FeSi showing that $S(Q,\omega)$ indeed exhibits a strong peak

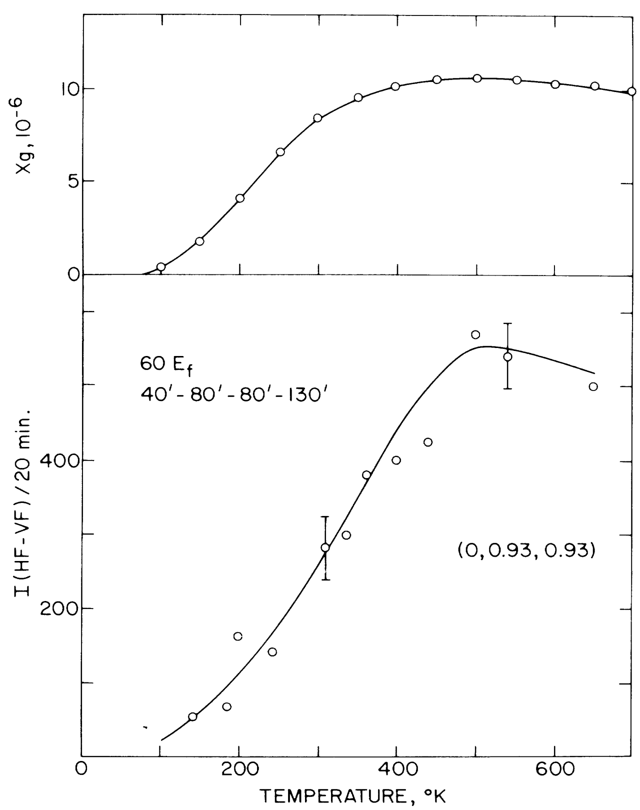

FIG. 1. Comparison of the magnetic susceptibility (Ref. 1) and the magnetic scattering of FeSi. The neutron data are taken near (011) with energy resolution FWHM of 15 meV. The line is a guide to the eye.

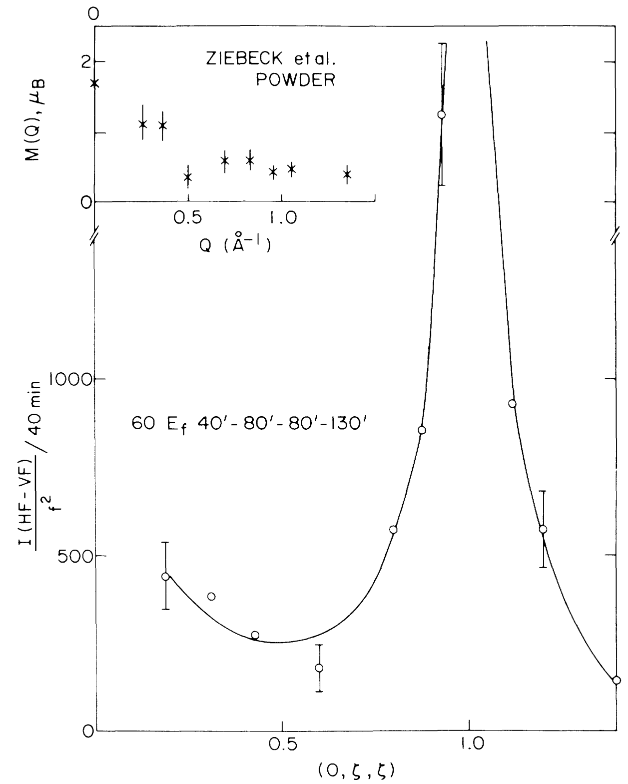

FIG. 2. The Q dependence of magnetic cross sections along the $[0,\zeta,\zeta]$ direction at 500 K, normalized by f^2. Inset: Powder data by Ziebeck *et al.* (Ref. 10). $d^*(011) = 1.98$ Å^{-1} at 300 K.

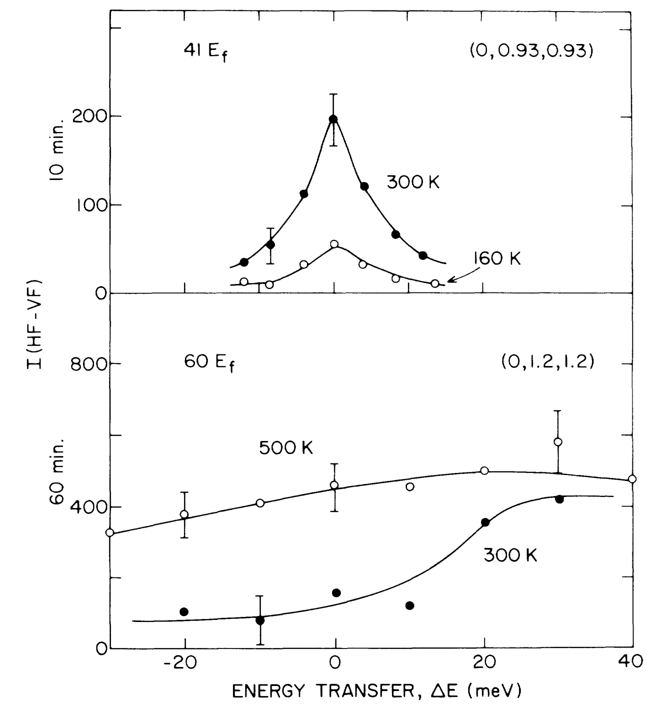

FIG. 3. Selected constant Q scans for FeSi. Collimation is 40′-80′-80′-130′. The data for (0,0.93,0.93) at 80 K are asymmetric. The data for (0,1.2,1.2) at 200 K lie below 100 at all energies. Lines are a guide to the eye.

at (011), implying ferromagnetic correlations, but the energy-integrated data show no q dependence. This novel combination of properties does not fit the usual picture of ferromagnetic correlations but is consistent with the model of temperature-induced paramagnetism. We studied a large (~ 12 cm^3), nearly perfect single crystal of FeSi recently grown at Tohoku University. Preliminary work on a smaller, less perfect specimen failed to show any correlations, partly because we *assumed* that the magnetic cross section was q independent and thus concentrated our measurements away from Bragg peaks to minimize background. With the new crystal it became feasible to explore systematically a wide range of q, T, and excitation energy. We used Heusler crystals as both polarizer and analyzer, and varied the energy transfer via E_i, the final energy E_f being fixed at 41 or 60 meV. We were obliged to use rather poor collimation, 40′-80′-80′-130′, to obtain sufficient intensity. Magnetic scattering was isolated by taking the intensity difference $I(\mathrm{HF}) - I(\mathrm{VF})$ between horizontal and vertical fields. Details may be found in Wicksted, Böni, and Shirane.[11]

As shown in the lower portion of Fig. 1, the small-q magnetic scattering intensity shows a temperature dependence which closely follows $\chi(T)$. In Fig. 2 we show the q dependence along $(0,\zeta,\zeta)$ of the magnetic scattering at 500 K, normalized to the form factor for comparison with Ref. 10. Our low-Q data suggest ferromagnetic correlations, consistent with Ref. 10 (inset), which are much more clearly evident in our data around the (011) Bragg point (corresponding to 2.0 Å^{-1}, a region that was not scanned in the powder experiment). We emphasize that the data shown in Figs. 1 and 2 are obtained with the analyzer set at $\Delta E = 0$ of 60-meV neutrons, with instrumental resolution (FWHM) of 15 meV.

At present we have limited data of $S(Q,\omega)$, as shown in Fig. 3, not enough to characterize the scattering function for the entire Brillouin zone. When we compare the top and bottom panels of Fig. 3, the energy width of the "correlations" increases rapidly with q as we go off the (011) Bragg point (0.07 top vs 0.2 bottom, in reduced units q). Furthermore, this energy broadening is markedly asymmetric at large q, especially at low temperatures. These observations indicate that neither of the data sets in Fig. 2 represents the q dependence of the magnetization $M(q)$. This can be obtained by energy integration of magnetic cross sections:

$$M^2(q) \propto \int [I_{\mathrm{mag}}(\mathrm{HF}) - I_{\mathrm{mag}}(\mathrm{VF})] d\omega,$$

from which we estimated $M(q)$ for the q and T range

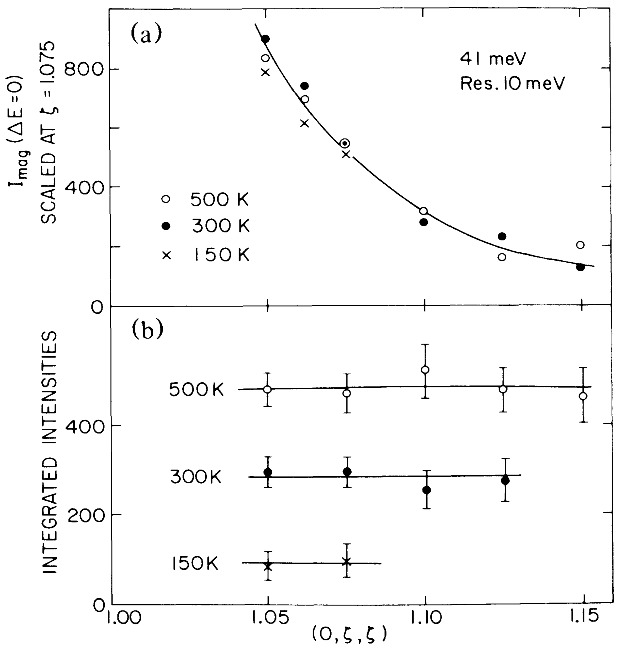

FIG. 4. (a) Normalized magnetic intensities for selected temperatures. (b) Energy-integrated magnetic cross sections as functions of momentum transfer q at selected temperatures.

where the observed profiles are symmetric. Surprisingly, these are nearly q independent for a wide temperature range (Fig. 4). At present there is no theoretical prediction of the unique characteristic of magnetic scattering from FeSi. Moreover, the shape of $I_{mag}(q)$ is nearly constant for a wide range of temperatures (see Fig. 4) and only the intensity increases. This increase, we believe, is the unique signature of temperature-induced magnetism in FeSi.

Now let us examine these neutron-scattering data from FeSi in the context of what we know about the general characteristics of paramagnetic scattering from a cubic ferromagnet above T_C. The scattering function is

$$S(Q,\omega)=M^2(0)\frac{\kappa_1^2}{\kappa_1^2+q^2}\frac{\Gamma}{\Gamma^2+\omega^2}\frac{\omega/kT}{1-e^{-\omega/kT}}, \quad (1)$$

$$M^2(q)=M^2(0)\frac{\kappa_1^2}{\kappa_1^2+q^2}, \quad (2)$$

with $Q=2\pi\tau+q$ and $\Gamma=Aq^{2.5}$ at T_C. The strong q dependence of magnetic scattering prevails near T_C where the inverse correlation length κ_1 is small. At high temperature the system becomes truly paramagnetic, namely $\kappa_1\rightarrow\infty$. However, the linewidth Γ remains q dependent and tends to approach zero as $q\rightarrow 0$. This was demonstrated in high-temperature studies[12] of the localized ferromagnetic Pd_2MnSn. At $T=4T_C$, this crystal shows a strong q dependence for Γ while $M(q)$ is almost q independent. The unique feature of FeSi is the strong temperature dependence of $M^2(0)$, which is not found in localized ferromagnets.

The shape of the scattering function $S(Q,\omega)$ is symmetric Lorentzian at high temperature, $kT>\Gamma$, because the temperature factor [last term in Eq. (1)] is nearly unity. At large q and low temperature, the observed shape is asymmetric, in particular, the 300 K data at (0,1.2,1.2) in Fig. 3. Lower-q data also exhibit similar asymmetry at lower temperatures. These line shapes are mainly caused by the thermal factor in Eq. (1) and probably not directly reflecting the energy gap in the system.

All features of magnetic scattering for FeSi presented in this Letter are consistent with temperature-induced magnetism in which magnetic electrons are thermally excited. In this sense, one should not use "ferromagnetic" correlations to describe FeSi. There are many important problems remaining for this unique magnet FeSi including more precise characterization of $S(Q,\omega)$, in particular at high- and low-temperature limits. More specific theoretical calculations will also be needed to design additional experiments to probe the model of temperature-induced magnetism in detail.

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[a]Permanent address: University of Pennsylvania, Philadelphia, PA 19104.

[1]Y. Ishikawa, Y. Noda, Y. J. Uemura, C. F. Majkrzak, and G. Shirane, Phys. Rev. B **31**, 5884 (1985), and references therein.

[2]T. Moriya, in *Spin Fluctuations in Itinerant Electron Magnetism,* Springer Series in Solid State Sciences Vol. 56, edited by M. Cardona, P. Fulde, H. J. Queisser, and K. von Klitzing (Springer-Verlag, Berlin, 1985).

[3]V. Jaccarino, G. K. Wertheim, J. H. Wernick, L. R. Walker, and Sigurds Arajs, Phys. Rev. **160**, 476 (1967).

[4]O. Nakanishi, A. Yanase, and A. Hasegawa, J. Magn. Magn. Mater. **15–18**, 879 (1980).

[5]Y. Takahashi and T. Moriya, J. Phys. Soc. Jpn. **46**, 1451 (1979).

[6]D. M. Edwards, J. Phys. F **12**, 1789 (1982); S. N. Evangelou and D. M. Edwards, J. Phys. C **16**, 2121 (1983).

[7]H. Watanabe, H. Yamamoto, and K. Ito, J. Phys. Soc. Jpn. **18**, 995 (1963).
[8]K. Motoya, M. Nishi, and Y. Ito, J. Phys. Soc. Jpn. **49**, 1931 (1980).
[9]M. Kohgi and Y. Ishikawa, Solid State Commun. **37**, 833 (1981).
[10]K. R. A. Ziebeck, H. Capellmann, P. J. Brown, and P. J. Webster, J. Magn. Magn. Mater. **36**, 160 (1983).
[11]J. P. Wiksted, P. Böni, and G. Shirane, Phys. Rev. B **30**, 3655 (1984).
[12]M. Kohgi, Y. Endoh, Y. Ishikawa, H. Yoshizawa, and G. Shirane, Phys. Rev. B **34**, 1762 (1986).