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# Inelastic neutron scattering studies of the paramagnetic phase in iron

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The paramagnetic phase of Fe(4%–Si) has been investigated by using polarized neutron scattering for temperatures ranging from the Curie temperature to $\sim 1.2T_c$ and for $q$'s ranging from 0.12 to 0.5 Å$^{-1}$ at the 011 reciprocal lattice point. All measurements were put on an absolute intensity scale via simultaneously measured phonon cross sections. Our energy resolved measurements $S(\vec{Q},\omega)$ consisted of a broad energy distribution centered at zero energy transfer. However, no spin-wave-like peak was present in any of these constant-$Q$ scans which contradicts the large "spin-wave" peak reported at $q = 0.47$ Å$^{-1}$ by Lynn. All recent theoretical models based upon this constant-$Q$ peak should be reexamined.

The investigations and their interpretations concerning the paramagnetic phase of bcc iron, as well as other ferromagnetic transition metals, have been the center of much controversy. The early inelastic neutron scattering studies by Collins *et al.*[1] observed critically damped modes at small-$q$ values above the Curie temperature. However, several years later, Lynn[2] reported the existence of spin waves well above $T_c$ for $q \geqslant 0.25$ Å$^{-1}$. Prange and Korenman,[3] Capellmann,[4] and Sokoloff[5] theoretically interpreted Lynn's results as arising from a large short-range magnetic order in iron's paramagnetic phase. However, this hypothesis was shown to be in conflict[6] with earlier specific-heat and paramagnetic susceptibility measurements made on iron.

Recently, Brown *et al.*[7] at Grenoble reported polarized neutron scattering measurements of the paramagnetic fluctuations in iron for temperatures ranging from $1.25T_c$ to $1.54T_c$ and for $Q$'s ranging from 0.23 Å$^{-1}$ to beyond the zone boundary along three principal directions. Using a poor energy resolution [full width at half maximum (FWHM) $\sim$43 meV], the Grenoble group interpreted their $S(\vec{Q})$ results as being in agreement with the theory of a large short-range magnetic order and concluded that ferromagnetic correlations of up to 15 Å existed well above $T_c$. However, it has been suggested[8] that for $q > 0.5$ Å$^{-1}$, the large neutron energy window used by Grenoble had not properly integrated the energy distribution $S(\vec{Q},\omega)$ of the paramagnetic scattering. Assuming that up to 70% of the scattering intensity was missing in the Grenbole data at $T = 1.25T_c$ and using a quantitative theoretical approach, Edwards[9] concluded that a very small correlation between nearest-neighbor spin directions existed in the paramagnetic phase of Fe.

One approach to help resolve the present controversy on the size of the short-range magnetic order above $T_c$ in paramagnetic iron is to utilize polarized neutrons along with an energy resolution which permits the magnetic scattering function $S(\vec{Q},\omega)$ to be properly observed. We have recently used such an approach to study the paramagnetic phase of $^{60}$Ni.[10] The constant $Q$ scans performed above $T_c$ ranging up to 0.4 Å$^{-1}$ in the [111] direction resulted in broad Lorentzian line shapes for the magnetic scattering with no indication of spin-wave peaks. These results in $^{60}$Ni motivated our present investigation of the paramagnetic phase in iron.

All polarized inelastic neutron scattering measurements were performed on a rectangular parallelpiped crystal of Fe(4%–Si) (4% at.wt) with dimensions $1\times1\times\frac{1}{4}$ in.[3] Triple axis scans in the [011] direction were made in the neighborhood of the 011 reciprocal-lattice point. The [0$\bar{1}$1] axis of the crystal was perpendicular to the horizontal scattering plane. Vertically magnetized Heusler alloy crystals were used as monochromators and analyzers along with magnetic guide fields which maintained the polarization of the neutrons. A 120-Oe guide field was imposed at the sample along the scattering vector. The focusing analyzer was fixed at 60 meV and all collimators were 40 min resulting in an energy resolution of $\sim$7 meV (FWHM) and a $q$ resolution of 0.07 Å$^{-1}$ for zero energy transfer at the 011 Bragg peak. A flat coil spin flipper placed between the sample and the Heusler analyzer was used to separate the spin-flip magnetic scattering (flipper ON) from the non-spin-flip nuclear scattering (flipper OFF). This separation was possible since the nuclear incoherent scattering from the sample was small while the instrumental flipping ratio was high ($R \approx 17$). All magnetic cross sections were put on to an absolute intensity scale based on a calibration curve resulting from theoretically calculated and experimentally measured phonon cross sections.

Figure 1 illustrates the scattering function $S(\vec{Q},\omega)$ for inelastic constant $Q$ scans of Fig.1(a) (0, 1.07, 1.07) ($q \approx 0.21$ Å$^{-1}$) at $T = 1.1T_c$, and Fig. 1(b) (0, 1.15, 1.15) ($q \approx 0.46$ Å$^{-1}$) at $T = 1.2T_c$ ($T_c \approx 1032$ K). The solid lines through the data points are guides to the eye for the magnetic scattering while the broken lines are similar guides for the nuclear scattering. The arrow in Fig. 1(b) corresponds to the expected spin-wave energy extracted from the

 

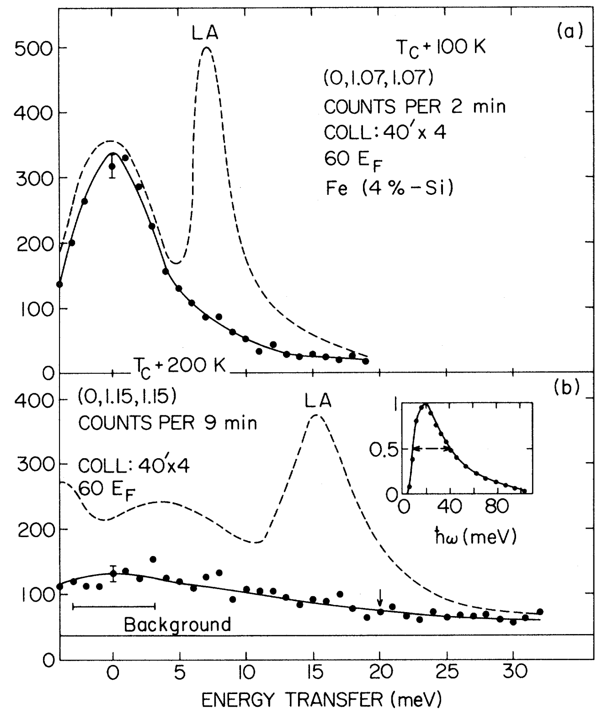


FIG. 1. Constant $Q$ scans of the paramagnetic scattering in Fe(4%–Si) at (a) (0, 1.07, 1.07) for $T=1.1T_c$ and (b) (0, 1.15, 1.15) for $T=1.2T_c$. The broken curve represents non-spin-flip scattering, while the solid line through the data points represents spin-flip scattering. The arrow points towards the expected spin-wave peak. Inset in (b) is copied from Fig. 10 of Ref. 2. The temperature-independent background (10 counts per 1000-K monitor) is shown in (b).

dispersion curve given in Ref. 2. The inset in Fig. 1(b) is just Fig. 10 of Ref. 2 which corresponds to a constant $Q$ scan on an Fe(12%–Si) crystal with reduced wave vector (0, 1.152, 1.152) ($q \approx 0.47$ Å$^{-1}$) and $T=1.28T_c$ ($T_c=970$ K). The magnetic $S(\vec{Q},\omega)$ data in Figs. 1(a) and 1(b) can simply be described as broad energy distributions of Lorentzian line shapes centered at zero energy transfer. Note, in particular, that no well-defined spin-wave peak is observed in our constant $Q$ data of Fig. 1(b) in contrast to the large spin-wave peak seen in the inset. This Lorentzian-like behavior was characteristic for all our $S(\vec{Q},\omega)$ measurements which ranged in $q$ from 0.12 to 0.5 Å$^{-1}$.

We have noticed in our constant $Q$ scans that for $(1+\zeta) \geqslant 1.1$, the "OFF" intensity shows a multiphonon scattering ridge between zero energy transfer and the LA phonon energy [see Fig. 1(b)]. This ridge is especially strong for studies using large neutron energies at high temperatures and thus may cause difficulty in the proper subtraction of a background for unpolarized neutron scattering studies. This difficulty suggests that Lynn's $^{54}$Fe(12%–Si) sample should be reexamined using polarized neutrons. Such a joint experiment is currently being planned with Lynn and Mook.

For each constant $Q$ scan, the energy integrated intensity $S(\vec{Q})$ of the magnetic scattering function $S(\vec{Q},\omega)$ was determined by integrating the broad energy distribution over the energy range covered. The value for $S(\vec{Q})$ was then converted into barns using the phonon calibration curve mentioned earlier after which it was converted into Bohr magneton squared units ($\mu_B^2$) and denoted by $M^2(\vec{Q})$. Figure 2 shows values for $M^2(\vec{Q})$ as a function of reciprocal-lattice units for four temperatures above $T_c$. Included in this figure are some of the results obtained by Brown *et al.* (open triangles) for measurements performed on Fe(5%–Si) at $T=1273$ K ($1.25T_c$) around the 110 Bragg peak. The solid lines drawn through the data points at different temperatures in Fig. 2 are guides to the eye. These lines begin at values of $M^2(0)$ which have been calculated from the static susceptibility.[11] The broken horizontal line in the figure shows the wave-vector-independent response expected for an ideal paramagnet with an effective magnetic moment $\mu_{\text{eff}}=3.12\mu_B$ calculated from the Curie constant. The $M^2(\vec{Q})$ data seen in this figure indicate a strong temperature dependence for $(1+\zeta)<1.1$ ($q<0.3$ Å$^{-1}$) after which this dependence decreases until the data at all temperatures measured converge to $\mu_{\text{eff}}^2$ for $(1+\zeta)\approx 1.15$. It is clear from this figure that our results from $M^2(\vec{Q})$ are in good agreement with the results obtained by Grenoble.

On the basis of the $M^2(Q)$ data obtained by Grenoble at $T\sim 1.25T_c$, Edwards quantitatively showed that if a large short-range order were present, the value of $M^2(Q)$ at $q\approx 0.1$ Å$^{-1}$ would become extremely large ($\sim 2025\mu_B^2$). We note that our value of $M^2(Q)$ at $(1+\zeta)=1.04$ ($q=0.12$ Å$^{-1}$) for $1.2T_c$ is much lower ($\sim 52\mu_B^2$) than the value calculated for giant short-range magnetic order by Edwards. On the basis of this result and our $S(Q,\omega)$ data, we feel that the theory of a large short-range order persisting well above $T_c$ in iron should be reexamined.

Brown *et al.*[7] have recently published two limited energy resolved (constant $Q$) scans which are similar to our results. However, they have claimed that "in both scans the quasielastic intensity is sufficient to dominate and obscure a small inelastic component." Clearly, our energy resolved results

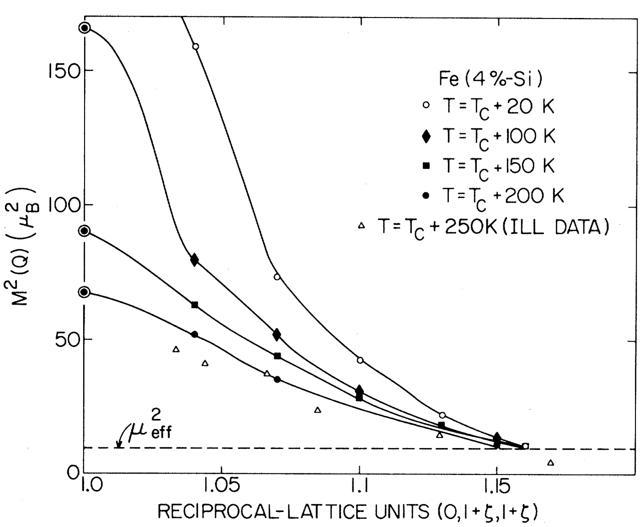


FIG. 2. Paramagnetic scattering $M^2(Q)$ in Fe(4%–Si) along the (0, $1+\zeta$, $1+\zeta$) direction for four temperatures above $T_c$. The solid lines through the data are guides to the eye for different temperatures. The values of these lines at $q=0$ have been calculated from the susceptibility. The broken horizontal line represents ideal paramagnetic scattering with an effective magnetic moment $\mu_{\text{eff}}=3.12\mu_B$. In addition, data obtained by the Grenoble group at $1.25T_c$ are denoted by open triangles.

in Fig. 1, which were performed with a better energy resolution, demonstrate that the cross section resembles a simple Lorentzian-like curve.

In summarizing our $S(\vec{Q},\omega)$ results, we have extended the small-$q$ investigations of Collins *et al.* using constant $Q$ scans for four temperatures above $T_c$ in Fe(4%–Si). In our measurements, we have not observed the constant $Q$ spin-wave peaks previously reported by Lynn for $q \geqslant 0.25$ Å$^{-1}$, although we have seen magnetic scattering ridges in constant energy scans which resemble the sloppy spin waves observed in similar scans conducted by Lynn. Our $M^2(\vec{Q})$ data agree with the data obtained by Grenoble although we feel the interpretation of these results should be reexamined. We believe that the magnetic scattering function $S(\vec{Q},\omega)$ has been adequately covered for small-$q$ values and small energy transfers. The next series of experiments on paramagnetic iron, which are currently being performed by us using polarized neutron scattering, are to investigate higher-$q$ values and larger energy transfers of $S(\vec{Q},\omega)$. All measurements of $S(\vec{Q},\omega)$ will be placed onto an absolute intensity scale with use of the technique described in this Rapid Communication allowing comparison with theories utilizing various sizes of short-range order.[3–6,12–14] We expect the constant $Q$ scans at these higher-$Q$ values to show a deviation in $S(\vec{Q},\omega)$ from the Lorentzian line shape illustrated in Fig. 1.

We have recently performed constant $Q$ polarized neutron experiments on pure iron at $1.02T_c$ ($T_c \approx 1044$ K). The magnetic scattering results are very similar to those obtained from Fe(4%–Si) at the same reduced temperature. This similarity seems to indicate that the silicon concentration in the Fe(4%–Si) has a negligible effect on the magnetic scattering in the paramagnetic phase.

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