Measurements of the magnetic excitations above T_c in iron and nickel (invited)

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High-resolution neutron scattering measurements, utilizing full polarization analysis techniques, have been carried out to explore the nature of the magnetic response above T_c in these itinerant ferromagnets. Large isotopically enriched single crystals were employed in order to obtain reliable determinations of the scattering function $S(q,E)$. The scattering in Ni is found to evolve in a continuous fashion from the well-known spin diffusion response at small wave vectors $\mathbf{q}$ to a response with peaks centered at $\pm E$ at larger $\mathbf{q}$, in agreement with our original work. The ratio of the observed widths of these spin waves to the spin-wave energies, as determined directly from measurements at constant $\mathbf{q}$, is found to decrease with increasing $|\mathbf{q}|$ as expected. We also show that the recent measurements by Shirane and collaborators were taken with resolution which was too coarse to observe these spin-wave excitations.

There continues to be great interest in the magnetism of the transition metals like nickel and iron because of the complicated nature of their magnetic interactions. Recently considerable attention has been focussed on the nature of the paramagnetic state of these systems, and some controversy has arisen both experimentally and theoretically concerning the nature of the spin dynamics above T_c. This paper serves to present the results of constant-$\mathbf{q}$ measurements with full polarization analysis which show that the scattering crosses over from spin diffusion behavior at small wave vectors $\mathbf{q}$ to spin-wave behavior at larger $|\mathbf{q}|$ as we concluded originally.[1-3] We also address the central issue of the experimental controversy which concerns the question of the appropriate instrumental resolution needed to obtain a reliable determination of the scattering function in this crossover region. The present measurements were taken with an overall resolution which is more than an order of magnitude better than that employed by Shirane and collaborators,[4,5] and we show that their poor resolution precluded them from observing any structure in $S(q,E)$.

EXPERIMENTAL CONSIDERATIONS

The measurements were taken with the HB-1 triple-axis polarized beam spectrometer at the High Flux Isotope Reactor at the Oak Ridge National Laboratory. The polarizer and analyzer crystals were both ^{57}Fe, which have good reflectivity and a d spacing of 2.02 Å, which provides inherently better resolution than the Heusler-alloy polarizers ($d = 3.445$ Å) more commonly used. Large isotopically enriched single crystals [^{60}Ni weighing 403 g and ^{54}Fe(12% Si) weighing 180 g] were used as samples in order to achieve an adequate signal-to-noise ratio while allowing us to employ sufficient instrumental resolution to determine the scattering reliably. Thermocouples were spot-welded onto the samples to measure the temperature, and the critical scattering and flipping ratio were monitored as a function of temperature to establish that the thermocouples were properly calibrated. A small field of 50 Oe was used to define the neutron polarization direction at the sample. The flipping ratio above T_c was about 20.

In order to appreciate the experimental difficulties inherent in making measurements on highly dispersive systems like the $3d$ transition metals, it is necessary to discuss some essential features of the neutron scattering technique and instrumental resolution. Figure 1 compares the low-temperature spin-wave disperison relations for Ni[1] and the Heisenberg ferromagnet EuO[6] with the free particle dispersion relation for neutrons. It is obvious that there is a better match for EuO than for Ni so that we might expect that measurements on an energy scale appropriate to Ni will be

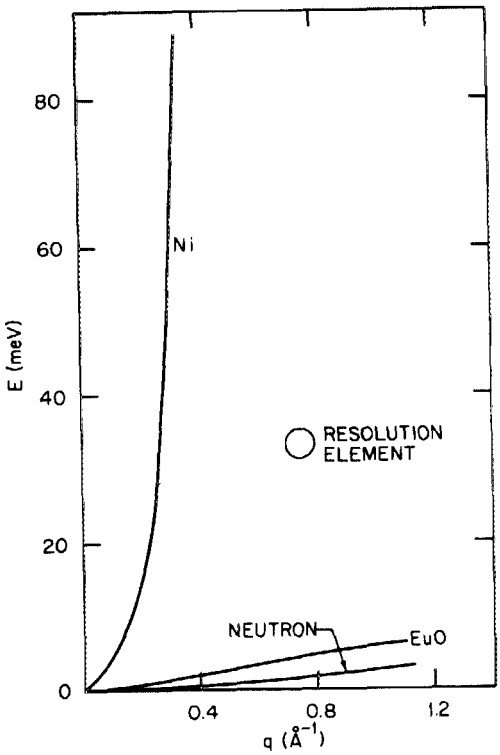

FIG. 1. Comparison of the dispersion relations for nickel, EuO, and the neutron. The circle represents a typical resolution element.

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difficult. The important point we want to make here is that the effects of instrumental resolution on the observations of spin waves in these two materials are fundamentally different. To appreciate this fact note the circle on the diagram, which represents a typical resolution element (0.1 Å^{-1} by 4 meV). Measurements are made by moving the element through the dispersion surface and observing the scattered intensity. For example, a constant-**q** scan is performed when the intensity is observed as a function of energy E at fixed **q**. If we make such a scan for EuO we see that the width of the observed scattering (assuming no intrinsic width for the spin wave) is dominated by the energy resolution. For nickel, on the other hand, a constant-**q** scan will give an observed peak which is very broad in energy since the resolution element will maintain "contact" with the dispersion curve over a large range in energy. Here the observed energy width is mainly determined by the wave vector resolution and the slope of the dipersion relation. In addition, the difficulties in interpreting such constant-**q** data are compounded by the fact that the actual resolution changes shape and volume rather dramatically over such a scan. Consequently the dispersion relations for such highly dispersive (i.e., large slope) systems are routinely established by performing constant-E scans in which the energy transfer is held constant and **q** is varied. The observed width is then narrow and is directly related to the wave vector resolution. In this case there is, of course, not one but three **q** components of resolution to consider.

Above T_c the scattering function is broadened, but it is still very dispersive[1-3] so that the constant-E technique is still preferred for obtaining accurate measurements of peak positions (q_0), widths, and shapes; in our opinion such data are the most reliable and detailed currently available to compare with theory. To determine if there are spin-wave excitations above T_c as evidenced by a peak in the scattering at finite energy, however, a constant-**q** scan would obviously be preferred *IF* sufficient resolution can be employed to obtain a reliable result. Table I gives a comparison of the instrumental resolution employed in the experiments of interest. The first line gives the values we employed in our original measurements with unpolarized neutrons. This serves as a bench mark by which to compare the recent polarized beam measurements. The resolution function is an ellipsoid in ($\mathbf{q},E$) space, and we have listed the projections along the three **q** axes (FWHM in Å^{-1}) and the energy axis (FWHM in meV). The last column gives a number which is directly proportional to the overall volume of the ellipsoid, and hence represents a figure of merit for these dispersive systems. The second line gives the resolution used in our present experiment with polarized neutrons. Note the overall resolution is a factor of 18 larger than we used originally. The reason for this is that the intensities achieved with polarized beams, although vastly improved over those available a decade ago, are still quite weak; typically there is a loss of $\sim 20\times$ in going to full polarization analysis. The coarsening of the resolution then compensates for the loss of spectrometer intensity. The advantage of the polarization technique is that it allows an unambiguous separation of the magnetic response from nuclear scattering (phonons, incoherent scattering, furnace and background scattering), which is a crucial consideration in the region of relatively small energies. The third line in the table gives our resolution for a fixed outgoing energy of $E_f = 60$ meV. Here the overall resolution is coarser, but there is an intensity increase since the region in ($\mathbf{q},E$) sampled by the spectrometer is much larger. Lines 4 and 5 show the resolution employed in the recent Brookhaven measurements. We see that their overall resolution is more than an order of magnitude coarser than that used for the present measurements, and between two and three orders of magnitude coarser than our unpolarized beam data. We will show as we present our data that the Brookhaven resolution is in fact too coarse to allow the observation of any structure in the scattering function. For completeness line 6 gives the resolution employed in recent measurements of Brown *et al.*[7] at ILL. We see that their resolution is comparable to that of Brookhaven.

RESULTS AND DISCUSSION

The general form for the scattering function $S(q,E)$ for an isotropic magnet can be written as[8]

$$S(q,E) \propto \chi(q) F(q,E) \frac{E/kT}{1-\exp(-E/kT)}. \tag{1}$$

Here $\chi(q)$ is the wave-vector-dependent susceptibility, which at small q is taken to be of the form

$$\chi(q) \propto \frac{1}{\kappa^2 + q^2}, \tag{2}$$

where κ is the inverse correlation range. The spectral weight function $F(q,E)$ contains the information about the shape of the scattering as a function of energy, and hence is the quantity of direct interest here. At small E ($E \ll kT$), where spin diffusion theory is valid, $F(q,E)$ is a Lorentzian centered at zero energy with a width $\Gamma(q)$ which is strongly q dependent:

$$F(q,E) = \frac{\Gamma(q)}{\pi[\Gamma(q)^2 + E^2]}. \tag{3}$$

For example, at $T = T_c$ we have $\Gamma = Aq^{5/2}$, with $A = 350$ meV $\text{Å}^{5/2}$ for Ni and $A = 3.3$ meV $\text{Å}^{5/2}$ for EuO. Thus the scattering for nickel is also very dispersive above T_c.

About twelve years ago, measurements were made by Mook *et al.*[1] which showed the then surprising result that the neutron scattering as determined by constant-E scans was rather similar above T_c to that found below T_c. In both cases a ridge of scattering was found that increased rapidly in energy from $q = 0$. This ridge softened as T approached T_c but was not strongly temperature dependent above T_c. We then showed[2] that the ridge broadened in q as the temperature increased, but the widths of the constant-E scans were still quite sharp above T_c. At small (q,E) these constant-E peaks could be understood on the basis of Eqs. (2) and (3), but with increasing q the spin diffusion formula gave increasingly poorer agreement with the observations. Indeed the widths became sufficiently narrow that we estimated $\Delta E/E < 1$ for energies above about 35 meV for Ni assuming that the ridge represented a dispersion curve. In this case the scattering

should not be thought of in terms of spin diffusion but rather in terms of damped propagating excitations.

Over the next few years measurements above T_c became available on other systems, and Fig. 2 shows the characteristic energetics for some of these materials. We note there is a close correspondence between the maximum (zone boundary) spin-wave energy for EuO,[6] EuS,[9] and Gd[10] and the thermal energy of the phase transition (kT_c). Above T_c constant-**q** scans for these materials show that the scattering has propagating character for q's near the zone boundary (~ 1.0 Å^{-1}), and these data are in reasonable accord with theory.[11] For a system like nickel, on the other hand, the spin-wave dispersion relation reaches kT_c at considerably smaller values of q, especially in comparison with the maximum allowed wave vector (2.0 Å^{-1}). Thus in analogy with materials like EuO and EuS, propagating character should be expected above T_c in the region where the excitation energies become comparable to kT_c. The unusual feature about nickel then is not that spin waves exist above T_c, but that T_c is so low compared to the characteristic energies of the magnetic excitations. From this view point the discovery of spin-wave excitations above T_c in the $3d$ transition elements for q values where the excitation energy becomes comparable to kT_c is not surprising but indeed is expected.

To obtain a clear picture of the magnetic scattering in the region where there are substantial contributions from the nuclear cross sections, it is highly desirable to use polarized-beam techniques as developed by Moon, Riste, and Koehler at Oak Ridge.[12] For the case when the neutron polarization $\hat{P} \| \mathbf{q}$ (horizontal configuration) all the magnetic scattering is spin-flip, while all the nuclear scattering is non-spin-flip. Simply measuring this cross section is often sufficient to determine the magnetic cross section, particularly if the beam polarization is high, the nuclear cross sections are relatively small, and the background has a small variation over the scan. This is the case for the measurements to be presented

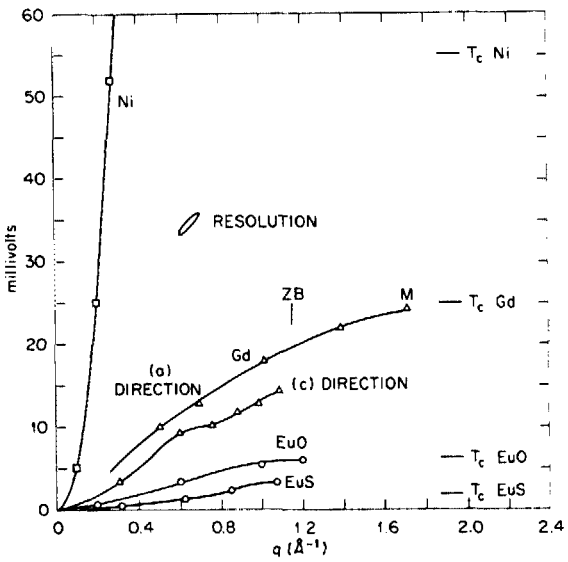

FIG. 2. Comparison of the ground state spin-wave energies for a number of ferromagnetic systems with the thermal energy needed to destroy long-range order.

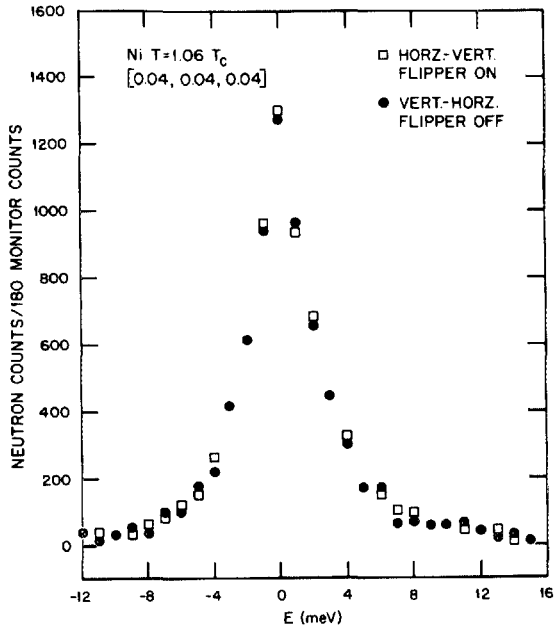

FIG. 3. Polarized beam measurements for nickel at small q, where spin diffusion theory is valid. The agreement between the two sets of subtracted data demonstate that the proper operation of the spectrometer was achieved.

on these isotope crystals. However, for $\hat{P} \perp \mathbf{q}$ (vertical configuration) half of the magnetic scattering is spin-flip and half is not, while the nuclear scattering is again non-spin-flip. Thus, the magnetic cross section can also be determined by subtracting $\hat{P} \perp \mathbf{q}$ from $\hat{P} \| \mathbf{q}$ in the spin-flip configuration, or by subtracting $\hat{P} \| \mathbf{q}$ from $\hat{P} \perp \mathbf{q}$ in the non-spin-flip configuration. This subtraction procedure has the advantage that the back-

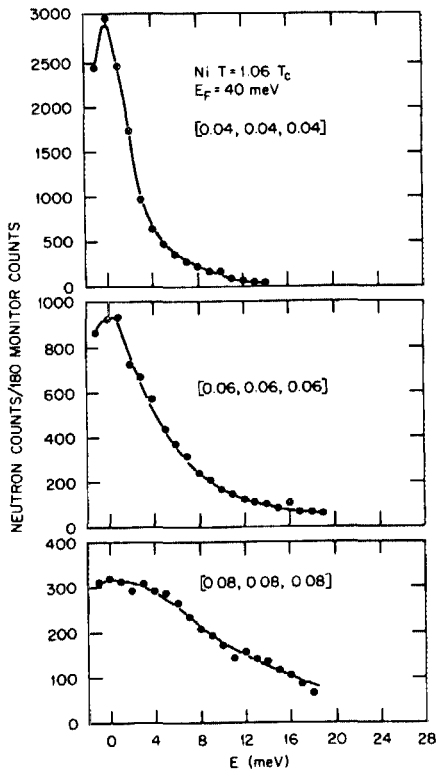

FIG. 4. Spin-flip (magnetic) scattering above T_c for a series of q's. The solid curves are fits to Lorentzians.

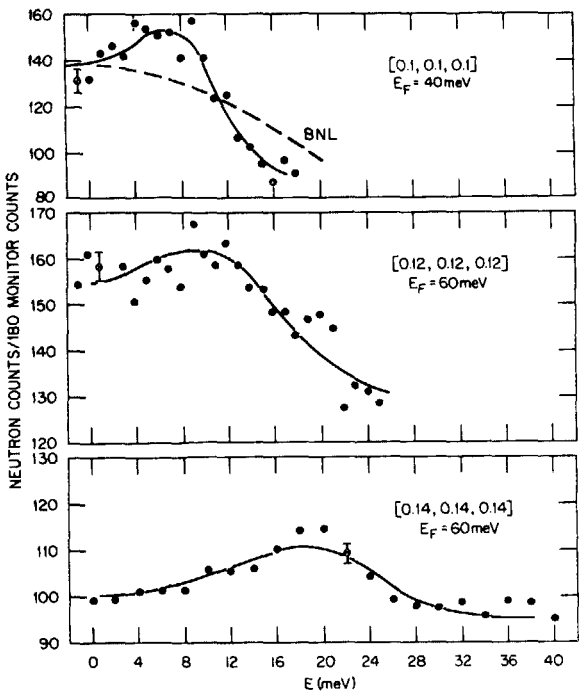

FIG. 5. Spin-flip scattering above T_c at larger values of q, showing that the scattering peaks at a finite energy rather than at $E = 0$. Thus the scattering in this region should be interpreted in terms of heavily damped spin waves rather than in terms of spin diffusion. Several runs were averaged together to obtain the data shown. Statistical uncertainty is shown by the error bars. The solid curves are the result of a least-squares fit to a damped harmonic oscillator form for the spectral weight function. The dashed curve is the expected scattering profile if the Brookhaven resolution is employed.

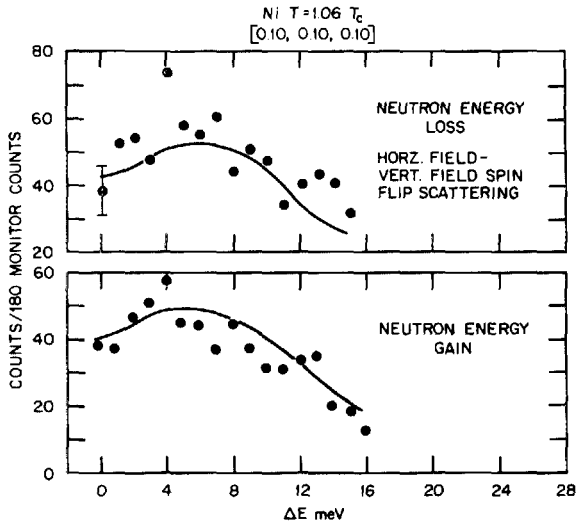

FIG. 6. Polarized beam data using the subtraction technique for both energy gain and energy loss. The data again are the average of several runs. The solid curves are the damped harmonic oscillator fits.

ground cancels, and hence any anomalous variations in the background cannot affect the result. Figure 3 shows the results of these subtractions for nickel at 1.06 T_c (669 K). At this small value of q ([0.04,0.04,0.04] in reduced units, which corresponds to 0.12 Å^{-1}) the scattering is diffusive, and the two subtraction procedures yield identical results demonstrating the high accuracy of our polarized beam data.

Figure 4 shows constant-**q** scans of the spin-flip scattering for larger values of q along the [111] direction. The scattering is seen to increase in width as expected, and the solid curves have been obtained by a least-squares fitting of a Lorentzian function to the data. Figure 5 shows the important results of this paper, where we see that with further increase of q the scattering develops structure and in fact no longer peaks at $E = 0$. The solid curves are least-squares fits to a damped harmonic oscillator form for the spectral weight function convoluted with the instrumental resolution function. The dashed curve for the [0.1,0.1,0.1] measurement is the result that would be obtained with the Brookhaven resolution using the identical harmonic oscillator parameters in the convolution. The coarse resolution is seen to wash out the structure in $S(q,E)$ in such a way that the observed scattering intensity would peak at $E = 0$. This is also the case (at this q) if we coarsen our own resolution by using an exit energy of 60 meV (Table I). However, the high-resolution data demonstrate that the scattering at this q is not diffusive in nature. Figure 6 shows the results of the subtraction technique for [0.1,0.1,0.1], and the results are seen to be in good agreement with the spin-flip data of Fig. 5. The solid curve is the damped harmonic oscillator fit we obtained using the same parameters as in Fig. 5.

The first appearance of spin-wave peaks at a finite $\pm E$ occurs at a q value of [0.1,0.1,0.1], which corresponds to 0.31 Å^{-1}. Thus, the crossover value q_c at this temperature must be at least this small. Our fits for the damped harmonic oscillator form for $F(q,E)$ give values of Γ/E of 1.12, 0.99, 0.69, 0.7, and 0.5 for $\xi = 0.06$, 0.08, 0.10, 0.12, and 0.14 in the $[\xi\xi\xi]$ direction. Thus, our best estimate for the crossover wave vector at this temperature is $q_c \sim 0.25$ Å^{-1}, which is in fact somewhat smaller than our original estimates.[1,2]

Figure 7 shows some results obtained on the ^{54}Fe(12% Si) single crystal above T_c at a reduced wave vector of

TABLE I. Resolution parameters (FWHM projections of resolution ellipsoid) for the experiments of interest assuming an energy transfer of 20 meV. The collimations are in minutes, the d spacings are in Å, the energies are in meV, and the wave vectors are in Å^{-1}. The smaller the volume the better the overall resolution figure of merit for the instrumental configuration employed.

Exp.	d	Coll.	E_f	Δq_x	Δq_y	Δq_z	ΔE	Vol.
ORNL[1-3]	1.7326	40-20-20-60	33	0.043	0.093	0.08	1.888	≡1.0
ORNL	2.0202	40-40-40-120	40	0.068	0.205	0.19	4.1666	18
ORNL	2.0202	40-40-40-120	60	0.080	0.296	0.23	7.034	63
BNL[4,5]	3.445	40-40-40-40	60	0.144	0.415	0.23	10.57	241
BNL[4,5]	3.445	40-80-80-80	60	0.190	0.717	0.23	17.42	904
ILL[7]	1.77	35-35-35-35	116	0.088	0.344	0.32	11.08	178

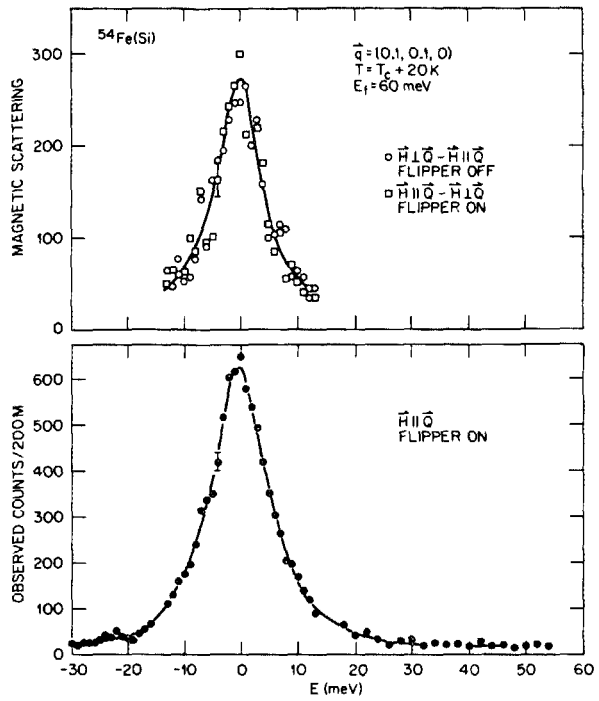

FIG. 7. Top part of the figure shows the polarized beam data for iron in the spin-diffusion region using the subtraction technique. The bottom part shows the spin-flip scattering at the same value of q, indicating our good signal-to-noise ratio. The nuclear (non-spin-flip) scattering is smaller than the magnetic signal over this entire scan.

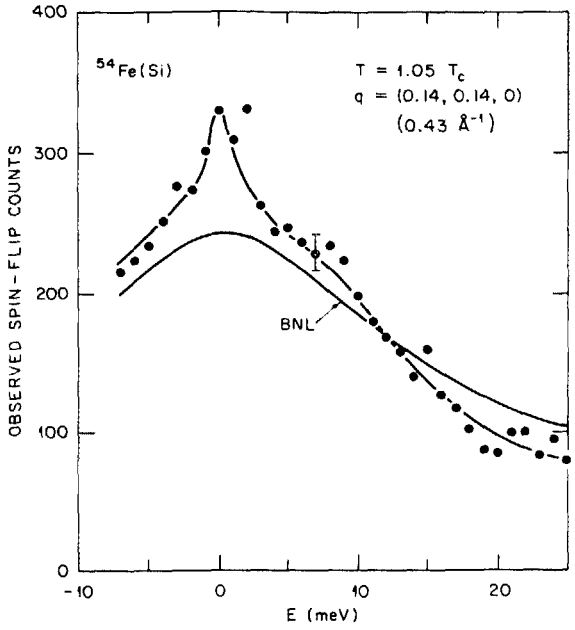

FIG. 8. Spin-flip scattering at a somewhat larger value of q, where structure is beginning to develop in the observed scattering. The solid curve is a fit to the damped harmonic oscillator form for the spectral weight, plus an elastic component. The curve labeled BNL shows how the identical cross section would appear using the resolution employed by Brookhaven.

[0.1,0.1,0] (0.31 Å^{-1}). The top part of the figure shows data similar to that for Ni given in Fig. 3, and demonstrates that the spectrometer was operating properly. The spin-flip scattering given in the lower portion of the figure indicates that a good signal-to-noise was achieved. The scattering is diffusive at this q, in agreement with our original data[3] as well as the data of Boronkay and Collins.[13]

At larger values of q we have obtained only preliminary results on iron, but they clearly show the development of structure in $F(q,E)$. Figure 8 displays the spin-flip scattering data at 1.05 T_c for $q = [0.14,0.14,0]$. Clearly, there are two components to the scattering, a quasielastic peak and broad shoulders. The solid curve through the points is a fit to a resolution-limited elastic component and a dampled harmonic oscillator spectral weight function. At this value of q(0.43 Å^{-1}) we obtain $\Gamma/E \approx 1.0$, so that we tentatively identify this as the crossover value. This value is in good agreement with our original results[3] as well as with our expectations based on the energetics of these materials.[8] We remark that the presence of the central component to the spin-flip spectrum, which broadens with increasing q, makes it very difficult experimentally to determine the crossover value for iron, and indeed complicates the interpretation of the cross section. It is important in this regard to establish whether the central component is an intrinsic property of pure iron, or whether it originates from the magnetic scattering associated with the silicon in the system. We do not observe a central component in nickel, and such a component has not been observed in related systems such as EuO, EuS, and Gd. Further measurements are now underway to better characterize the nature of the magnetic excitations of iron.

The curve marked BNL in Fig. 8 shows how the data would appear using the resolution in the Brookhaven experiments. This curve was obtained by convoluting the cross section we determined with the resolution shown in Table I (line 4). Clearly high resolution is needed to obtain a correct understanding of the magnetic excitations in these systems.

In summary we have directly observed in the paramagnetic state of Ni crossover from spin diffusive behavior at small wave vectors to progagating behavior at larger wave vectors. This has been demonstrated experimentally in a clear manner as we find scattering profiles for temperatures above T_c that are not centered at $E = 0$ but rather have finite excitation energies. This crossover occurs at 0.25 Å^{-1} for nickel, and preliminary data suggest $q_c \sim 0.43$ Å^{-1} for iron, which is in agreement with our esarlier estimates based on constant-E scans. We suggest this crossover behavior is determined by the value of the characteristic excitation energy in comparison to the transition temperature. In systems like iron and nickel the excitations at large q are at high energies and hence will not be dramatically affected by the thermal fluctuations with energy kT_c.

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