Observation of itinerant-electron effects on the magnetic excitations of iron

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Triple-axis neutron spectroscopy has been used to study high-energy magnetic excitations for iron. The scattering contours established for the [100] and [111] directions show effects not present in localized-electron models of ferromagnetism. Interactions between spin waves and single-particle excitations are clearly seen for both crystallographic directions while two parts to the spin-wave branch are observed for the [100] direction.

The magnetism of iron has been the subject of numerous investigations over many years. We know that the electrons responsible for the magnetism of iron are at least partly itinerant as they contribute to the low-temperature heat capacity, and the Bohr-magneton number for iron is not an integral number. However, the magnetic-excitation spectrum at low energies is similar to that expected for a Heisenberg ferromagnet, and indeed at one time, this was used as an argument that a localized-electron model was appropriate for iron. We know now that a band model which properly takes into account the electron correlations produces Heisenberg-like spin waves at low energies and in addition, predicts itinerant-electron effects at high energies. In this paper we show high-energy neutron-scattering measurements that clearly demonstrate itinerant-electron effects for the first time.

Measurements of high-energy magnetic excitations were made with the hot-source triple-axis spectrometer at the Institut Laue-Langevin using a large (200 g) single-crystal sample of ^{54}Fe alloyed with about 10% silicon. The low nuclear cross section of ^{54}Fe is very helpful in reducing nonmagnetic scattering events, and the silicon is needed to stabilize the body-centered phase for iron. The silicon is not expected to alter the nature of the magnetic excitations very much, other than to reduce their energy scale. The spin-wave stiffness is, in fact, reduced about 15% by the addition of the silicon.[1] A vertically focusing Cu monochromator set for reflection by the (331) planes and a focusing Cu analyzer utilizing the (220) reflection were used in the experiment. Collimation of 40′ was used after the monochromator, and 60′ collimation was used before and after the analyzer. All measurements were made using a constant analyzer energy of 364.9 meV so that an In filter could be used to eliminate $\lambda/2$ contamination. A Sm filter was used in the incident beam so that low-energy thermal neutrons that were scattered off the monochromator by incoherent and inelastic processes were absorbed and did not reach the sample. The sample was maintained at a temperature of 77 K to reduce phonon scattering.

The magnetic excitations were measured at energy transfers as high as 350 meV requiring incoming neutrons with energies of near 700 meV. We found that the hot source provided sufficient numbers of neutrons of this energy and the main difficulty in the experiment was that very small scattering angles were needed to simultaneously satisfy the energy and momentum conservation relations in the experiment. Since the neutron background was angle-sensitive at small scattering angles, measurements were generally made with and without the sample and the difference taken. This procedure yielded results that were free of background variations over the course of the scans.

Measurements were first made for the [100] direction. The data were generally accumulated by making constant energy scans at about 5 meV intervals over the region of interest. The scans showed interesting results as the sharp spin-wave excitations found at low energies were found to broaden considerably at about 150 meV and then to become narrow again at higher energies. Previous triple-axis[1,2] measurements have been made at energies up to 120 meV and an experiment using a pulsed source obtained some data at energies as high as 160 meV.[3] The new measurements are consistent with the spin-wave stiffness established in the earlier results and with the fact that the spin-wave intensities diminished in the neighborhood of 100 meV. The pulsed-source measurements were analyzed assuming an isotropic dispersion surface which we will see is far from the case. However, if results for the various directions in the crystal are averaged together, a result similar to the time-of-flight measurement would be obtained.

We were guided in our measurements by the calculations by Blackman *et al.*[4] Indeed their calculation showed that for the [100] direction, a sharp spin wave was to be expected at low energies which faded out in the neighborhood of 200 meV, but that an additional spin-wave branch becomes prominent at high energies. Because of the high energies used in the experiment, spectrometer resolution effects were important, and it was necessary to take them into account. Generally in analyzing the triple-axis results, a trial dispersion surface is convoluted with the four-dimensional (three wave vector and one energy) spectrometer resolution and the result is least squares fitted with the data. Parameters are then adjusted for the dispersion surface until a good fit is obtained. The analysis thus proceeded by assuming a single isotropic dispersion surface, convoluting this with the resolution, and adjusting the slope, width, and intensity of the dispersion surface until the best possible agreement with the data was obtained. This does not give a very good representation of the data, and the band calculation suggested that the scattering would be better represented by using two independent dispersion surfaces. The data were thus reanalyzed using two dispersion surfaces and separate parameters for the position, width, and intensity of the two surfaces. One of the surfaces established the lower-energy spin waves while the other determined the high-energy branch. This gave a better fit to the data as might be expected because more parameters were used. Figure 1 shows three scans for the [100] direction where the solid curve is the fit using the two-dispersion-surface model. The spin waves at 52 and 248 meV are nearly resolution-limited while the result for 155 meV is much broader than the resolution.

There is no way to tell convincingly from the present data whether more than one dispersion surface really exists. In any case, the result of interest is the position of the scattering contours, and these are shown in Fig. 2 as determined from the two-dispersion-surface model. This result is entirely inconsistent with the scattering as calculated by a localized-electron model but closely resembles that obtained by the band calculations of Blackman *et al.*[4] The rapid broadening and decrease in intensity of the spin wave near 150 meV is an itinerant-electron effect. The sharp mode that is found to begin at around 200 meV and extends out of the energy range of the measurement shows that the spin-wave dispersion curve for the [100] direction appears to consist of more than one part. The results near 300 meV are the highest spin-wave energies attained in any measurement, and the scattering contours in Fig. 2 give a clear indication of the unusual behavior of the excitations of a material whose electrons are at least partly itinerant.

Figure 3 shows results obtained for the [111] direction. In this case, only one dispersion surface is needed to give a good fit to the data. The spin waves start out as sharp excitations and broaden considerably as they near the

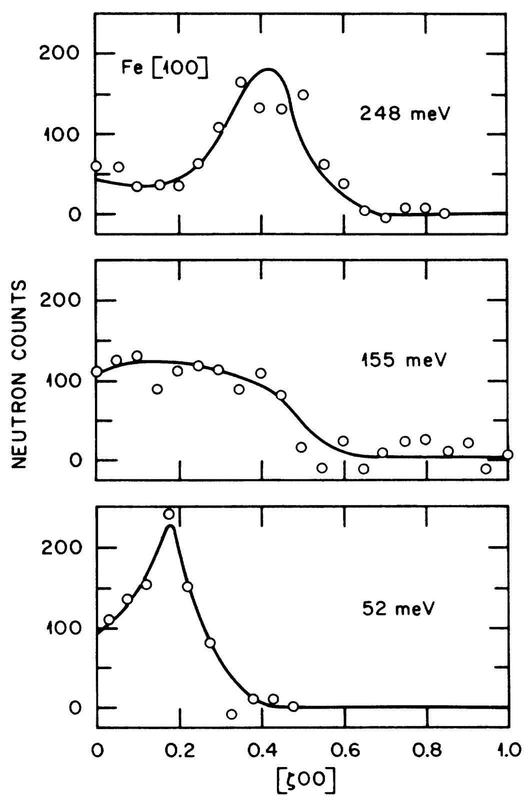

FIG. 1. Triple-axis measurements of spin waves for Fe for the [100] direction. The solid line is the least-squares fit using two isotropic dispersion surfaces.

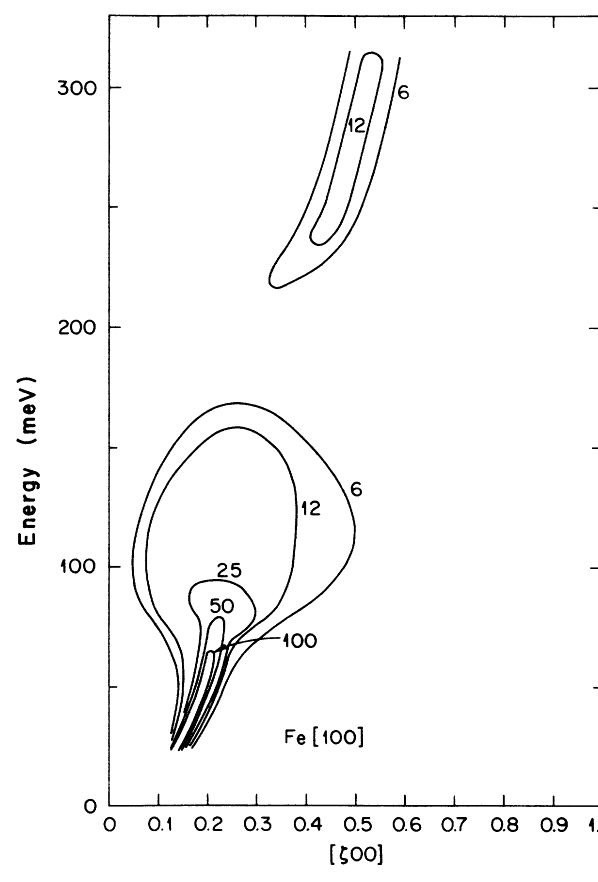

FIG. 2. Scattering contours for the [100] direction for Fe obtained by least-squares fitting of the two-dispersion-surface model, convoluted with the spectrometer resolution, to the data.

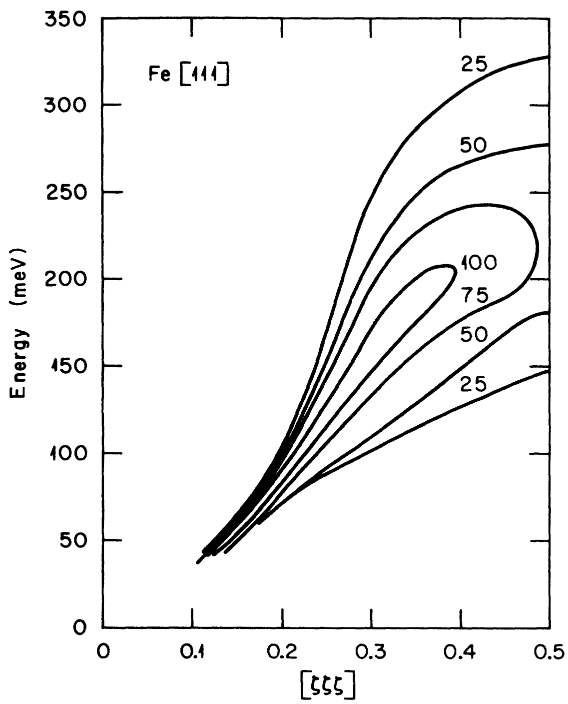

FIG. 3. Scattering contours for the [111] direction for Fe obtained by least-squares fitting of a model dispersion surface, convoluted with the spectrometer resolution, to the data.

zone boundary. The broadening takes place when the spin waves encounter a dense region of single-particle excitations, and this again is an itinerant-electron effect. Note that the complete dispersion surface has been established for the [111] direction.

Before a close comparison can be made between the band calculations and the measured result, there is an additional resolution effect that must be considered. The data were analyzed in terms of isotropic dispersion surfaces, and as we see, the results for the [100] and [111] direction are very different. The spectrometer resolution extends off symmetry so that some sampling of the dispersion surface between symmetry directions is included in the measurements. It is too costly in spectrometer time to measure in the off-symmetry directions, so some way must be established of estimating the error made by the off-symmetry resolution effects. This was accomplished by convoluting a nonisotropic dispersion surface with the spectrometer resolution and examining the results. A single dispersion surface was chosen that fits the high energies observed for the [100] direction and the lower energies for the [111] direction, gradually varying in between. The result of convoluting the spectrometer resolution with this surface was that the energies were lowered by about 6% for $\mathbf{q}=0.3$ in the [100] direction. The energies for the [111] direction increased slightly. Obviously, if the dispersion surface lowers rapidly in going off symmetry from the [100] direction, a larger error is made. A few off-symmetry band calculations have been made for the regions near the [100] direction which suggest that the fall off is not extremely rapid.

A comparison between the measured results and the band calculations for the [111] direction shows excellent agreement in both the position and width of the scattering contours. The position is expected to be raised slightly by the off-symmetry resolution effects and lowered slightly by the silicon in the sample so that the effects may nearly cancel. The agreement for the [100] direction is not as good. The lower-energy part of the dispersion surface is steeper than that given by the calculation suggesting some improvement in the calculation may be needed. The broadening of the excitations in the region near 150 meV is correctly provided by the calculation. The measured higher-energy part of the spin-wave branch is lower than that given by the calculation but both the silicon and off-symmetry resolution effects are expected to lower the experimentally determined energy contours. Over all, the agreement between the band calculation and measurement is surprisingly good considering the difficulty of both the measurement and the calculations. Similar results have previously been observed for nickel[5] although the measurements were not nearly so comprehensive and contour maps were not established for the spin wave intensities. Clearly, it would be beneficial to extend the measurements in both Fe and Ni to higher energies and to off-symmetry directions. This may be possible if high-equality beryllium monochromators can be developed for reactor instrumentation or if the new pulsed neutron sources will have sufficient intensity.

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