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High-energy spin waves in bcc iron

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The dispersion relation of the spin-wave excitations in bcc iron has been studied by neutron inelastic scattering at the spallation neutron source ISIS. Magnetic intensity was followed up to 550 meV along the [100] direction. The general form of the dispersion curve is in qualitative agreement with that calculated from a spin-polarized band model, and in particular the prediction of propagating modes extending above 300 meV has been confirmed.

I. INTRODUCTION

A complete description of the static and dynamic properties of magnetic systems is contained in the generalized magnetic susceptibility $\chi(\mathbf{q},\omega)$ which can be calculated from a model of the spin-polarized band structure of a system. The neutron-scattering cross section is directly related to the imaginary part of the generalized susceptibility, and so neutron-scattering measurements of the wave vector $\mathbf{q}$ and frequency ω dependence of the magnetic excitations can be used to provide a sensitive test of calculations of $\chi(\mathbf{q},\omega)$ from the band theory.

Various experiments have shown that the $3d$ electrons in iron occupy narrow bands that are itinerant in character. At energies of less than about 100 meV the excitations in iron at room temperature consist of sharp spin waves isotropic in $\mathbf{q}$ and with almost constant intensity.[1,2] Such behavior is consistent with the picture of a localized, Heisenberg ferromagnet. $\chi(\mathbf{q},\omega)$ is much more sensitive to itinerant effects at high energies, where the collective modes can interact with single-particle excitations in the Stoner continuum, and significant deviations from the localized model were observed during a recent neutron-scattering study of iron[3] in which the scattering intensity contours were mapped out to energies of 300 meV in the [100] and [111] directions. Above 100 meV the spin waves were observed to broaden considerably, decrease in intensity, and become anisotropic in $\mathbf{q}$, and in the [100] direction the spin-wave branch was found to split into two parts. These observations agree very well with the predictions from a calculation based on a spin-polarized band model,[4] which showed that the upper, "optic," branch of the dispersion relation is derived from interband transitions.

II. EXPERIMENT

The Oak Ridge single crystal of ^{54}Fe is approximately 170 g in mass and contains about 12% Si to stabilize the bcc phase. The spin-wave stiffness at low energies has been found to decrease by about 15%, and the Curie temperature by 7%, on addition of the Si,[5] but otherwise the magnetic properties are similar to elemental Fe. The isotope ^{54}Fe was chosen because of its small nuclear cross section that considerably reduces the nonmagnetic scattering. The sample was mounted with [001] vertical on a goniometer, and measurements were made at room temperature.

There are considerable experimental difficulties in measuring spin waves at these relatively large energy transfers. High-incident neutron energies must be combined with low scattering angles in order to satisfy the neutron energy and momentum conservation laws and at the same time to restrict the wave-vector transfer to values where the magnetic form factor is not too prohibitively small. The previous high-energy experiment was performed on the IN1 triple-axis spectrometer at the Institut Laue-Langevin, where the flux of high-energy neutrons is enhanced by means of a hot moderator. The present study was made at the spallation neutron source, ISIS, at the Rutherford Appleton Laboratory, on the high-energy-transfer spectrometer (HET) which derives a large flux of epithermal neutrons with intrinsically good time structure from an ambient temperature moderator. HET is a direct-geometry, time-of-flight spectrometer[6] that uses a rotating Fermi chopper phased to the source proton pulse to select the incident neutron energy with a resolution of between 1% and 2%, rather better than that of IN1. The scattered neutrons are detected in two banks of ^{3}He detectors situated at 4 and 2.5 m from the sample and covering the angular ranges 3°–7° and 9°–29°, respectively. The data presented here were taken in the horizontal arm of the 4-m bank which was in most runs masked down in height from 30 to 15 cm to improve the vertical resolution. A similar experiment[7] was performed on the chopper spectrometer LRMECS at the spallation source IPNS, Argonne Na-

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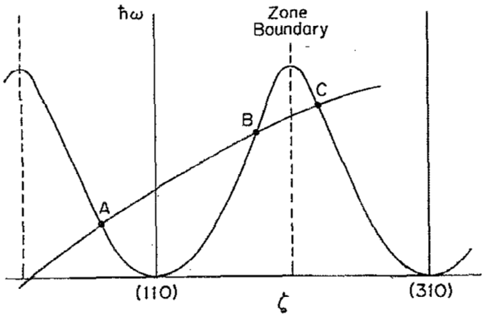

FIG. 1. Schematic representation of a time-of-flight scan parallel to [ζ00]. The locus of neutron energy transfers in a detector intersects the spin-wave dispersion surface in both the (110) and (310) zones, and the intersection points at A, B, and C result in peaks in the inelastic scattering.

tional Laboratory, in which spin waves were measured in pure iron up to 160 meV. The scans, however, were in no particular symmetry direction, and the resolution and intensity was much poorer than on HET.

Our procedure for measuring collective modes is different from that generally used on a reactor source. Whereas on a triple-axis spectrometer scans are usually performed with **q** along a symmetry direction and in constant-**q** or constant-ω mode, the time-of-flight method employs a constant scattering angle so that both the energy transfer and **q** vary with the scattered neutron's time of flight. To measure spin waves along a symmetry direction we aim to find a combination of incident neutron energy and crystal orientation so that the time-of-flight trajectory in reciprocal space is parallel to the symmetry direction and intersects the dispersion surface at the required energies. However, the incident energy in such a scan is not always a sensible choice; for example, it may be excessively large. Alternatively, we use advance knowledge to position the crystal so that the time-of-flight trajectory is not parallel to the symmetry direction except at the point of intersection with the dispersion surface. Figure 1 illustrates how a typical time-of-flight scan parallel to [100] intersects the spin-wave dispersion curve in different zones.

III. RESULTS

In this work we report on the measurement of spin waves along the [100] direction. A variety of incident energies and crystal orientations were used to ensure that the scans included spin-wave excitations in the [100] direction. In most cases the spin waves were measured about the (110) reciprocal lattice point from intersections of type A and B in Fig. 1, but the highest-energy excitations were obtained from type-C intersections in the (200) or (310) zones.

Because of the nature of the time-of-flight scan the magnetic excitations occur on a sloping, nonmagnetic background that arises from multiphonon processes. This background falls rapidly with increasing energy transfer and becomes negligibly small at an energy transfer roughly equal to half of the incident energy. Thus, for the measurement of weak magnetic signals at high energies the incident energy must be restricted to less than twice the excitation energy. To obtain reliable estimates of the peak positions it is essential to subtract the sloping background from the magnetic scattering. It was noted that the spectra in the 2.5-m detectors retained approximately the same shape and intensity across the whole angular range of the bank and, after normalization for solid angle by scattering from vanadium, gave a good estimate of the nonmagnetic background in the 4-m detectors.

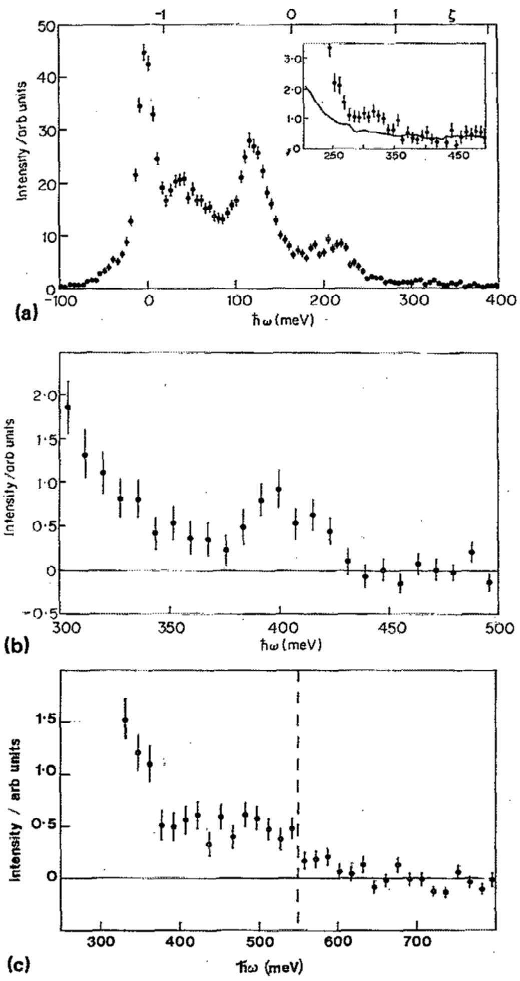

FIG. 2. Examples of spin-wave scattering from Fe(12%Si). ζ is the reduced scattering vector parallel to [100] measured from the (110) zone center. (a) Raw data for an incident energy of 600 meV without background subtraction. The inset shows the third spin wave in the scan, at 320 meV, together with the multiphonon background (the intensity below 250 meV is from the spin wave at 220 meV). (b) Spin-wave scattering at 400 meV on the upper, "optic," branch of the dispersion curve, measured with neutrons of incident energy 900 meV. The multiphonon background has been subtracted from the magnetic scattering. (c) Residual magnetic intensity in the region of the zone boundary (indicated by the vertical line) after background subtraction. The detectors were masked in (a) and (c), but not in (b).

As an illustration of the raw data Fig. 2(a) shows the observed scattering converted from time of flight to energy transfer for an incident energy of 600 meV before background subtraction. The spectrum has been corrected for the energy dependence of the detector efficiency and for the k_f/k_i phase-space term in the cross section, but not for the

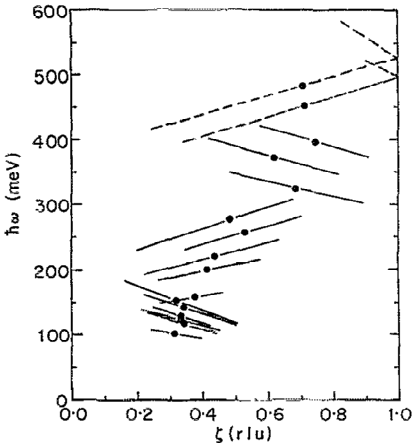

FIG. 3. Spin-wave dispersion curve of Fe(12%Si) at room temperature in the [100] direction. The bars are drawn parallel to the scan direction and correspond to the full width at half maximum.

magnetic form factor. Two spin waves can be resolved, at energies of 120 and 220 meV, and the second one may be seen to be broader than the first. These two peaks correspond to cuts of types A and B in Fig. 1. The inset to Fig. 2(a) shows that a weak third peak is present at 320 meV with low intensity. This is identified with a cut of type C to the dispersion surface in the (310) zone. Figure 2(b) shows the high-energy part of the spectrum measured with 900-meV incident neutrons and corrected for background as described above. The highest incident energy employed in this experiment was 1200 meV, and in Fig. 2(c) we focus again on the high-energy part of the data. For the crystal orientation used in this scan the time-of-flight trajectory intersects the zone boundary at an energy transfer of 550 meV, and the presence between 400 and 600 meV of residual magnetic intensity after background subtraction shows that we are close to the maximum excitation energy in Fe(12%Si).

IV. DISCUSSION

We present in Fig. 3 the spin-wave dispersion curve of Fe(12%Si) in the [100] direction derived from this experiment. The peak positions and widths have been obtained from the background-subtracted spectra without regard for resolution effects, so the data must necessarily be treated as preliminary. Nevertheless, the dispersion curve is not inconsistent with that derived from the band-structure calculation[4] and is in broad agreement with the previous study.[3] However no division into "acoustic" and "optic" branches is apparent at this level of analysis. We have measured the upper "optic" branch to an energy of 550 meV and the data indicate that the threshold of magnetic intensity at the zone boundary is of the order of 600 meV. With reference to type-C spin waves, we find the peaks to be particularly well defined in the vicinity of 400 meV and scans here both with and without detector masks appear to be greater in intensity than lower down on the "optic" branch. Generally speaking, the energy scale of the measured excitations is somewhat lower than predicted, and this may be due in part to the presence of Si in the sample. Through the superior resolution and background on HET the quality of the high-energy data obtained from the spallation source is better than that from reactor sources in the past, and we note that the spin waves measured in this study are considerably higher in energy than have ever been observed before.

A thorough analysis of the data is in progress at present to take into account the effects of resolution, and we aim ultimately to produce a contour map of the scattering intensities. We also plan high-energy measurements along other symmetry directions to extend our knowledge of $\chi(\mathbf{q},\omega)$ throughout more of the Brillouin zone, in particular the [110] direction in which $\chi(\mathbf{q},\omega)$ is especially sensitive to details of the band calculation. This further work will facilitate a more exacting test of the predictive power of the band theory of magnetism.

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