SPIN-WAVE AND CRITICAL SCATTERING OF NEUTRONS FROM COBALT DISULFIDE

M. Iizumi* and J. W. Lynn
Brookhaven National Laboratory*, Upton, New York 11973
A. Ohsawa
Tohoku University, Sendai, Japan
H. Ito
Hokkaido University, Sapporo, Japan

ABSTRACT

The triple-axis neutron scattering technique has been used to measure the temperature dependence of the spin-wave and critical scattering below \( T_c \) (120 K) in the metallic ferromagnet CoS\(_2\). The spin-wave dispersion relations at small propagation vectors \(|q| < 0.3 \text{Å}^{-1}\) can be described by \( \tilde{E}(q) = Dq^2 - E \) with \( D \approx 110 \text{ meVÅ}^2 \) and \( E/D \approx 1.2 \text{Å}^2 \) at 78 K, with a small anisotropy observed in the different symmetry directions of the crystal. At small \( q \), \( D \) is found to renormalize as \( (1 - T/T_c)^\beta \) with \( \beta = 0.24 \). Some indication of a triple-peaked structure in the scattering spectrum was observed immediately below \( T_c \).

The properties of the metallic conductor CoS\(_2\) and its solid solutions with FeS\(_2\) and NiS\(_2\) are considerably less complicated to interpret than the 3-d ferromagnetic elements and alloys because the former's electrical and magnetic characteristics both arise from the same single d-band which is free from hybridization with s or p bands. For this reason they have been extensively studied in recent years\(^1\) and many of their properties have been successfully interpreted in terms of the tight-binding Hubbard model. These systems therefore appear to be favorable to study the influence of the itinerancy of the d-electrons on the magnetic excitation spectrum.

In the present study we report measurements of the spin-wave spectrum and critical scattering for CoS\(_2\), which has the pyrite crystal structure and is ferromagnetic below 120 K.\(^1\) The spin-wave dispersion relations were measured on a single crystal which was grown by the chemical transport technique. Unfortunately, the neutron scattering measurements are hindered by the relatively large absorption and incoherent scattering cross sections for cobalt. Moreover, the magnetic moment is only 0.84 \( \mu_B \) per cobalt atom, and this coupled with the small sample size (0.2 cm\(^3\)) available has limited the present spin-wave measurements to wave vectors less than 0.3 \text{Å}^{-1}. Since the spin-wave dispersion was found to be approximately isotropic in \( \tilde{q} \), however, the...
measurements of the critical scattering and the renormalization of the spin waves at small wave vectors could be taken in the forward direction on a powder sample of considerably larger volume (5 cm$^3$). This sample was in the form of a plate 5 mm thick.

The neutron scattering measurements were made with the triple-axis spectrometers at the Brookhaven High Flux Beam Reactor. Most of the spin-wave measurements on the single crystal were taken with the constant-$E$ technique with an incident neutron energy of 41 meV. Because of the inherently weak inelastic intensity, we were obliged to use the relaxed horizontal collimation of 40$'$ full width at half maximum (FWHM) both before and after the pyrolytic graphite (PG) monochromator and analyzer. The nominal FWHM energy resolution under these conditions was 4 meV. The measurements on the powder were made with the constant-$Q$ technique. The incident energy in this case was 13.7 meV and 10$'$ collimation was used before and after the monochromator and analyzer. The nominal FWHM energy resolution was 0.2 meV, and useful data could be obtained for scattering angles larger than 0.5$'$. A PG filter was used for both 41 and 13.7 mev incident neutrons to reduce unwanted higher order wavelengths. All the spin-wave energies presented have been corrected for the finite resolution of the instrument.

The dispersion relations for the spin waves in the high-symmetry directions at 78 K are shown in Fig. 1. Our data extend only to 0.3 A$^{-1}$, which is about 1/4 of the way to the zone boundary (1.14 A$^{-1}$) in the [001] direction. Over this wave-vector range the spin-wave dispersion relation can be expanded in a power series in $q^2$. The data have therefore been least-squares fitted to the expression $\omega = Dq^2 - E q^4$. The resultant fits are shown in the figure as solid curves. For the three high-symmetry directions [001], [110], and [111], we obtained values for the spin-wave stiffness parameter $D$ of 105, 116 and 118 meV-A$^2$, respectively. The smaller value of $D$ in the [001] direction is outside our experimental error, so there appears to be a small amount of anisotropy present. On the other hand, measurements of the bulk magnetization show only a very small anisotropy. The ratios $E/D$ of the fourth-order coefficients to the second-order coefficients are 1.9, 1.6, and 1.1 A$^2$ for the [001], [110], and [111] directions, respectively.

The temperature dependence of the spin-wave scattering was observed in two different energy regions. For energies between 4 and 8 meV the magnetic scattering as measured in constant-$E$ scans shows well-defined peaks at low temperatures, which then increase in width with increasing temperature without any significant shift of the peak positions. Although a very broad
peak is observed even above $T_c$, one cannot attribute a propagating character to this peak, because the width of the peak suggests that there are no side peaks in the spectral weight function. In order to determine if there is any resemblance to the propagating behavior observed in iron and nickel, it is necessary to measure the magnetic scattering at higher energies and wave vectors, which are unfortunately experimentally inaccessible at present because of the small sample size.

At small values of $q$ the spin-wave energies change rapidly near $T_c$. It is convenient to present the renormalization of the spin-wave energies by the temperature dependence of the stiffness parameter $D(T)$. The results are shown in Fig. 2. We have fit these data to the expression $D(T) = D(0) (1 - T/T_c)^\beta$. With $T_c$ fixed to the measured value, the least-squares fit gave a value of $\beta = 0.24 \pm 0.02$, which agrees well with the exponent of $0.25 \pm 0.01$ obtained by Jibu et al. from measurements of the magnetization.

The observed temperature dependence of the magnetic scattering below $T_c$ is shown in Fig. 3 for a wave vector of 0.06 Å⁻¹. To obtain this data a large background correction has been made to the data at energies around $E=0$. This background originates primarily from air scattering and small angle scattering from the cryostat and sample. The observed scattering around $E=0$ at 78 K and room temperature coincided within experimental error and were taken as background. This amounted to $1245 \pm 15$ counts/23 minutes at $E=0$. The absence of a central component at 110 K assures that the background subtraction has been performed satisfactorily. The spectra at temperatures closer to $T_c$ suggest that there may be a triple-peaked structure, the two side bands being the spin waves at $\pm \omega_q$, and the central peak being due to longitudinal correlations. Such a central diffusive mode is not present in other nearly isotropic ferromagnets such as Fe, Ni, Co, EuS or EuO, whereas this component has been clearly seen in isotropic antiferromagnets such as PbMnF$_3$ and in very anisotropic ferromagnets such as MnF$_2$. Although the present data suggest that there is a central mode in CoS$_2$, clearly more effort will be required in order to unambiguously decide this question.

The authors are indebted to G. Shirane for suggesting this study and for many stimulating discussions. They have also benefited greatly by enlightening discussions with S. Ogawa, Y. Yamaguchi, and L. P assail. We would also like to thank C. J. Glinka for kindly providing us with one of the data processing programs.
REFERENCES

* On leave from Japan Atomic Energy Research Institute, Tokai, Japan.
† Work at Brookhaven performed under the auspices of the U. S. Energy Research and Development Administration.


Fig. 1. Dispersion relations along the principal symmetry directions of the crystal. Open circles indicate the data obtained with the single crystal sample, while the open squares show the data obtained with the powder sample, for which the isotropy of the dispersion relations at small wave vectors has been assumed. The solid curves are the best fits to the data for each direction. For comparison the best-fit [111] curve is shown as dashed curves in the [110] and [001] directions.
Fig. 2. Renormalization of the spin-wave stiffness parameter $D$ with temperature. The solid curve is the best fit to the data.
Fig. 3. Constant-Q profiles of the magnetic scattering below $T_c$ (122.5 K). Background has been subtracted. The data suggest that there may be a central component to the scattering below $T_c$. 