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TITLE THE COMMENSURATE SPIN EXCITATION IN CHROMIUM: A POLARISED
NEUTRON INVESTIGATION

AUTHOR(S) R. Pynn, W. G. Stirling, and A. Severing

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

**The Commensurate Spin Excitation in Chromium:
a Polarised Neutron Investigation**

R. Pynn

The Manuel Lujan Jr. Neutron Scattering Center, Los Alamos National
Laboratory, Los Alamos, NM 87545, USA

W.G. Stirling

Department of Physics, University of Keele, Keele, Staffordshire, ST5 5BG,
U.K.

and

A. Severing

Institut Laue Langevin, 156 X, F-38032 Grenoble, France

Abstract

A polarised neutron experiment with neutron energy analysis has been performed with a single-Q sample of chromium in a large magnetic field. The 4-meV "commensurate" mode is found to involve spin fluctuations parallel to the ordered chromium moments.

Running Title: Polarised neutron investigation of Chromium

Keywords: Polarised neutrons, inelastic scattering, chromium

Pure chromium is one of the most fascinating magnetic elements [1]. Below its Neel temperature, $T_N = 311\text{K}$, chromium has a sinusoidally modulated antiferromagnetic structure with a modulation vector $Q_0 = (1-x) G_{100}$, where x is approximately 0.05 and G_{100} is the $(1,0,0)$ reciprocal lattice vector. Usually, magnetic satellites are observed at all wavevectors that are crystallographically equivalent to Q_0 , but a single- Q state can be obtained by field-cooling a single-crystal sample in a magnetic field parallel to one of the $\langle 1,0,0 \rangle$ cube edges. Between T_N and the "spin flip" transition at $T_{SF} = 121\text{K}$, in the *transverse spin density wave* (TSDW) phase, the magnetic moments lie along $\langle 0,1,0 \rangle$ and $\langle 0,0,1 \rangle$ directions for the single- Q state in which Q_0 is parallel to G_{100} . Below T_{SF} in the *longitudinal spin density wave* (LSDW) phase, the moments are parallel to Q_0 .

The magnetic excitation spectrum of chromium exhibits several unusual features. In addition to the extremely steep "spin waves" that originate from the satellite positions, there are lower energy excitations that have been observed in the TSDW phase. Fincher et al [2] were the first to observe such an excitation and reported a peak in the fluctuation spectrum at 4 meV, centred on the commensurate wavevector G_{100} . This excitation is unaffected by an applied magnetic field [3] and disappears as T_{SF} is approached [2]. Studies of this commensurate excitation by Burke et al [4] indicated that the excitation split into two branches above 4 meV. Since these branches had the same slope as longitudinal phonons, Burke et al suggested that they might be due to longitudinal phonons observed via the magnetic interaction between neutrons and chromium ions, that is, magneto-vibrational scattering. This explanation is quite different from that suggested by Fincher et al [2] who proposed that the 4-meV peak could result from an excitation of the TSDW in which blocks of spins oscillate in the plane perpendicular to the TSDW wavevector, Q_0 .

A suitable polarised neutron experiment should be able to distinguish between the two explanations proposed for the low-energy magnetic excitations close to G_{100} . If a sufficiently large field is applied along the $\langle 0,0,1 \rangle$ direction in a single- Q , TSDW phase in which Q_0 is parallel to G_{100} , the preferred spin domain involves chromium spins that are parallel or antiparallel to the $\langle 0,1,0 \rangle$ axis [5]. In this situation, the twisting-block model proposed by Fincher et al involves magnetic fluctuations that are essentially parallel to $\langle 0,0,1 \rangle$ and these should give rise to neutron scattering that is non-spin-flip (NSF) because the quantisation direction for neutron spins is defined by the applied magnetic field to be along $\langle 0,0,1 \rangle$. On the other hand, since the magneto-vibrational scattering involves coupling to the ordered chromium moments that are directed principally along $\langle 0,1,0 \rangle$, longitudinal phonons seen through the magnetic interaction between neutrons and chromium moments should give mostly spin-flip (SF) scattering. An experiment to determine the nature of the low-energy magnetic excitations should therefore give a straightforward answer, supporting one or other of the proposed models. Such an experiment has been carried out using the IN20 triple axis spectrometer at the Institut Laue-Langevin. As so often happens, the results of this experiment are not as simple as one might have hoped (or feared!). Rather they demonstrate that chromium has not yet revealed all of its secrets.

The 1.5 cm^3 single crystal sample examined in this experiment was the one used for previous measurements by Burke et al [4] and ourselves [6]. It had been cooled through T_N in a large applied magnetic field to induce a 99% single- Q

state. The sample was mounted in a cryomagnet that could provide a field of up to 6 Tesla perpendicular to the (0,0,1) scattering plane of the experiment (see Figure 1 for the experimental configuration). Focussing Heusler alloy monochromator and analyser crystals were used to provide polarised neutrons. The spectrometer was operated with a fixed final neutron energy of 14.7 meV and a pyrolytic graphite filter was used to remove higher order contamination of the scattered neutron beam. A dc spin flipper was mounted in the scattered beam and, even when the largest fields were applied to the sample, a flip ratio of about ten was maintained. The results have been corrected for the effect of this flip ratio.

Werner et al [5] have already shown that the extent to which chromium moments can be forced into the (0,0,1) plane by a magnetic field applied along (0,0,1) depends on the quality of the chromium crystal used. Our sample was grown by iodine vapour transport and had a relatively small mosaic spread (20'). At the maximum field available to us (6T), we could only obtain a ratio of order 5 between the spin-flip and non-spin-flip intensities of the $Q = (0.954, 0, 0)$ TSDW satellite at a temperature of 150 K (see Fig. 2). This ratio falls to a value of about 3.5 at 230 K. Although the ratios we have measured are similar to those reported by Werner et al [5], the fields we used to obtain them were much higher. We attribute this to existence of more high-energy pinning sites in our sample. Without an accurate model for the orientation distribution of the chromium spins it is not possible to correct our data for the effect of pinning sites and we have not attempted to do so. The large magnetic field applied in this experiment had no effect on the single-Q nature of the sample, a fact that was confirmed from time to time during the measurements by observing the intensity of the (0,0.954,0) satellite.

Our experiment provided a number of surprises, as indicated by the data in figure 3. Although the commensurate excitation observed by Fincher et al [2] appears in the SF channel we do not believe that it is magneto-vibrational scattering. This conclusion is based on three facts:

- (a) we have been unable to observe magneto-vibrational scattering by transverse phonons in spite of the fact that such scattering should be as strong as magneto-vibrational scattering from longitudinal phonons;
- (b) the intensity of the 4 meV peak decreases by a factor of about 3.5 when the temperature is lowered from 230K to 150K. The decrease of the Bose factor and the increase of the TSDW satellite intensity almost cancel over this temperature range, so that a much smaller drop would be expected if the peak were due to phonons;
- (c) branches of excitations are observed above 4 meV but not below, even though a magneto-vibrational mechanism calls for intensity to increase as energy transfer decreases.

We are thus led to conclude that the commensurate feature at 4 meV is an excitation of the TSDW, even though it cannot be the "twisting block" excitation suggested by Fincher et al [2] because it appears in the SF channel. The excitation involves fluctuations of the chromium moments in the $\langle 0,1,0 \rangle$ direction, that is, parallel to the ordered TSDW moments. A scan at an energy transfer of 6 meV shows that the excitation in the SF channel splits into two peaks, as indicated by Burke et al [4]. However, since there

is no evidence for well-defined *SF* peaks in constant energy scans with energy transfers less than 4 meV, the dispersion relation for this branch would appear to have a minimum at 4 meV and $Q = Q_{100}$. There is *SF* intensity below 4 meV at the commensurate position, indicating perhaps that the 4 meV excitation is coupled to a diffusive dissipation channel. Our best guess at this stage is that the 4 meV excitation could represent a fluctuation of the amplitude of the TSDW.

The twisting-block fluctuation may be manifested by the intensity in figure 3 around the commensurate position in the *NSF* channel. There is certainly more weight in the *NSF* fluctuation spectrum at this point than can be explained by a simple overlap of the spin wave peaks that occur at the satellite wavevectors. However, if one makes the simplest possible correction for the existence of spin domains that are not parallel to $\langle 0,1,0 \rangle$, the *NSF* intensity at the commensurate position disappears, making firm conclusions impossible. We have some evidence that at 270K the *NSF* intensity at Q_{100} and 4 meV may give rise to a well defined peak as the Neel temperature is approached, but confirmation of this point requires further experimentation.

Another surprising feature of the data displayed in figure 3 is the intensity of the peaks at the satellite positions in the *SF* channel. In spite of the existence of spin domains not polarised along $\langle 0,1,0 \rangle$, a large part of this scattering corresponds to longitudinal magnetic fluctuations (i.e. fluctuations parallel to the ordered moment) and is remarkably large at a temperature that is already 80K below T_N (230K for figure 3). Previous work with localised [7] and itinerant ferromagnets [8] has shown that diffusive longitudinal critical fluctuations do exist and that they peak strongly at the paramagnetic-to-ferromagnetic transition. While one would expect similar critical scattering in an antiferromagnet, one would not expect it to be similar in magnitude to the spin-wave scattering 80K below the Neel temperature. Another remarkable feature is that the *SF* scattering at the satellite wavevectors has been found to extend over a very large energy range. Quite clearly, what was once thought to be spin-wave scattering in the TSDW phase, involves a substantial contribution from the longitudinal magnetic susceptibility. It is tempting to conjecture that the *SF* component of the scattering at the satellite wavevectors corresponds to phase fluctuations of the TSDW. However, more work is required before such an assertion can be made with any confidence.

While the results we have obtained have succeeded in overthrowing previous models for the excitations in the TSDW phase of chromium, they do not yet provide a complete alternative picture. More measurements are planned to establish the behaviour of both $\langle 0,0,1 \rangle$ and $\langle 0,1,0 \rangle$ fluctuations as T_N and T_F are approached.

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Figure Captions

- Figure 1: Relative orientations of chromium moments and spin density wavevector Q_0 in the scattering plane of the experiment. The magnetic field was applied perpendicular to the scattering plane. Satellite positions are indicated by full circles.
- Figure 2: Spin-flip and non-spin-flip intensities for the (0.954,0,0) satellite at a temperature of 150K as a function of applied magnetic field.
- Figure 3: SF and NSF intensities for a constant energy scan at 4 meV along the $\langle 100 \rangle$ direction at a temperature of 230K and applied magnetic field of 6 Tesla.