Characterization of low-energy magnetic excitations in chromium

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The low-energy excitations of Cr, i.e. the Fincher-Burke (FB) modes, have been investigated in the transversely polarized spin-density-wave phase by inelastic neutron scattering using a single-$\mathbf{Q}_\perp$ crystal with a propagation vector $\mathbf{Q}_\perp$ parallel to [0, 0, 1]. The constant-momentum-transfer scans show that the energy spectra consist of two components, namely dispersive FB modes and an almost energy-independent cross section. Most remarkably, we find that the spectrum of the FB modes exhibits one peak at 140 K near $\mathbf{Q} = (0, 0, 0.98)$ and two peaks near $\mathbf{Q} = (0, 0, 1.02)$, respectively. This is surprising because Cr crystallizes in a centro-symmetric bcc structure. The asymmetry of those energy spectra decreases with increasing temperature. In addition, the observed magnetic peak intensity is independent of $\mathbf{Q}$ suggesting a transfer of spectral-weight between the upper and lower FB modes. The energy-independent cross section is localized only between the incommensurate peaks and develops rapidly with increasing temperature.

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INTRODUCTION

Although chromium exhibits a simple bcc structure and consists only of a single element the magnetism is very complicated and one of the most intriguing subjects in condensed matter physics.\textsuperscript{1} Below the Néel temperature $T_N = 311$ K, the magnetic structure exhibits an incommensurate antiferromagnetic transverse spin-density-wave (TSDW) with the moments oriented perpendicular to the ordering wavevectors $\mathbf{Q}_\perp$. The pitch of the modulation is approximately $\mathbf{Q}_\perp \sim (0, 0, 0.045)$. Below the spin-flop temperature $T_{sf} = 122$ K the moments arrange along $\mathbf{Q}_\perp$ in a longitudinal spin-density-wave (LSDW). The pitch of the modulation is approximately $\mathbf{a}/\delta \sim 20\mathbf{a}$ at low temperatures.

The inelastic magnetic cross section shows also a surprisingly rich behavior (Fig.\textsuperscript{1}). High-velocity excitations (often called spin waves) emerge from the incommensurate positions $\mathbf{Q}_\perp$. The excitations exhibit longitudinal and transverse polarization, the former one being assigned to a phason mode. In particular, the longitudinal mode dominates below an energy transfer $E \approx 8$ meV, the incommensurate excitations become isotropic above 8 meV. These modes are present in the LSDW as well as in the TSDW phase.

In addition to the high-energy excitations at $\mathbf{Q}_\perp$, Fincher et al., Burke et al.\textsuperscript{2, 3} and Sternlieb et al.\textsuperscript{11} found another magnetic mode in the TSDW phase at low energies that is located between the incommensurate peaks. The dispersion relation of this so-called Fincher-Burke (FB) mode was reported to emanate from the $\mathbf{Q}_\perp$ positions and to merge into the high-energy excitations near $E \approx 8$ meV (Fig.\textsuperscript{1}). The two FB-branches cross each other at $E_c \sim 4$ meV and $\mathbf{Q}_c = (0, 0, 1)$. (Hereafter, we redefine the two branches as upper and lower FB mode, respectively, with the boundary energy of $E_c$.)

By means of neutron scattering with polarized neutrons it was shown that the FB modes are longitudinally polarized\textsuperscript{2, 11, 12}.

Very recently, Hiraka et al. re-investigated the FB mode using unpolarized neutrons and found by means of

FIG. 1: Dispersion of magnetic excitations based on the previous experimental results in the TSDW phase of Cr. The vertical cones show the dispersion of the high-energy excitations emerging from the incommensurate positions. The chain lines indicate the FB modes and are the subject of the present investigation. The scan trajectories are represented by horizontal and vertical lines. The ellipsoid indicates a typical resolution of the current thermal-neutron experiments.
constant-momentum-transfer scans (constant-Q scans) a new excitation mode \[Q_c \neq 0, E_c] that emerges from the point \((Q_c, E_c)\) towards a direction transverse to \(Q\) within the \((h, k, l)\) plane. Interestingly, the gap-type mode extends with a low velocity up to at least \(h, k = \pm 2\delta\) and shows a decreasing intensity with increasing \(h, k\). Using a cold neutron triple-axis spectrometer with high-resolution Böni et al. investigated the FB mode in detail [14] using constant-Q and constant-energy-transfer scans (constant-E scans). They pointed out that the FB mode neither shows a simple linear dispersion nor obeys a simple spin-wave picture with respect to intensity. In addition, the intensity contour for \(Q_- < Q < Q_+\) indicated that the FB mode is asymmetric with respect to \(Q_c\). However, it was not clear if this was due to spurious scattering.

In order to obtain a coherent picture of the low-energy excitations in Cr we have performed a detailed investigation of the FB modes in the TSDW phase by means of constant-Q scans using thermal- and cold-neutron spectrometers. Previous measurements were mostly performed by constant-E scans (Fig. 1). However, the intense high-energy excitations made the quantitative analysis of the weak response of the FB modes difficult.

The major result of this work is a confirmation of the asymmetry of the energy spectra with respect to \(Q_c\) near \(T_{sf}\) using a different sample and different spectrometers. We also observed that the energy spectra become more symmetric at high temperatures. In addition, we show that the FB modes conserve intensity along \(Q_{\pm}\) (or \(l\)), in contrast to the transverse mode that shows a drastic depression of intensity along \(h\) and \(k\) [13]. The nearly energy-independent component shows a strong \(T\)-dependence of the cross section that may be closely related to the commensurate scattering [3, 6] or the critical scattering discussed by Sternlieb et al. [10].

**EXPERIMENTAL PROCEDURE**

For the present experiments we have used the identical single crystal of \(\approx 4 \text{ cm}^3\) that was previously investigated [13]. The single-Q structure pointing along \([0, 0, 1]\) was induced by cooling the sample through \(T_N\) in a strong magnetic field yielding a domain population \(\geq 99\%\). The crystal was mounted inside a closed cycle refrigerator with the \((h, 0, l)\) scattering plane horizontal. The measurements with thermal and cold neutrons were conducted on the triple-axis spectrometer TOPAN installed at the JRR-3M research reactor at the Japan Atomic Energy Research Institute and the triple-axis spectrometer SPINS at the National Institute for Standard and Technology, respectively. The \((0, 0, 2)\) reflection of pyrolytic graphite was used to monochromate and analyze the neutron energy. The final neutron energy of TOPAN (SPINS) was fixed at 14.7 meV (3.7 meV), and the horizontal-collimation sequence was set to 30’-60’-30’-60’ (guide-40’-40’-open) from before the monochromator to after the analyzer. The energy resolution of the spectrometers was estimated to be less than 1.2 meV and 0.4 meV, respectively. Higher-order neutrons were removed by means of a pyrolytic graphite filter and a BeO filter for TOPAN and SPINS, respectively.

**EXPERIMENTAL RESULTS**

In Fig. 2 we show the salient results of the present work. It is clearly seen that spectra evolve from a single peak at \((0, 0, 0.97)\) towards a double-peak structure at \((0, 0, 1.02)\). The significance of this result was confirmed by reproducing the scans under different experimental conditions.

**FIG. 2:** Energy spectra measured along \([0, 0, 1]\) in the TSDW phase. A single peak and a double peak are observed at the symmetric \(Q\)-positions \((0, 0, 0.98)\) and \((0, 0, 1.02)\), respectively. The solid lines are drawn as a guide to the eyes.
FIG. 3: Energy spectra measured above and below $T_{sf} = 122$ K at (a) $(0,0,1)$ and (b) $(0,0,1.02)$. $I_{UFB(LFB)}$ represents a peak intensity of the upper (lower) FB mode above the $E$-independent cross section $I_{const}$ (broken line) at 140 K. The chain line indicates the non-magnetic background. The energy resolution is shown with horizontal bars.

Figure 3 shows detailed measurements at the commensurate position $(0,0,1)$ and at the intermediate position $(0,0,1.02)$ for temperatures below and above $T_{sf}$. The so-called Fincher peak at $(0,0,1)$ is broader than the $E$-resolution. A broadening of the peaks is also noticed in the scan at $(0,0,1.02)$. Below $T_{sf}$, the peak structure disappears completely. At the same time the $E$-independent magnetic scattering decreases too, but it is significantly larger than the background of 30 ~ 40 counts/5 min that was measured at various points in the Brillouin zone. The behavior of the $E$-independent component is consistent with “commensurate scattering” which has already been observed before by means of constant-$E$ scans using unpolarized and polarized neutrons.

In order to parametrize the data we have fitted the peaks in Figs. 2a and 3a after subtraction of the background to one or two Lorentzians plus a constant, $I_{const}$. We express peak heights of the former part as $I_{UFB}$ and $I_{LFB}$ for the upper and lower FB mode, respectively (Fig. 3). The latter $I_{const}$ is considered to be independent of $E$ but depends on $T$. Figure 4(a) summarizes the $Q$-dependence of the energy position of the FB mode. The vertical thick bars indicate the width of the peaks which are, roughly speaking, constant against $Q$. It is clearly seen that the dispersion is extremely asymmetric with respect to $Q_c$ because no peaks are observed at the low-$Q$ side of the lower FB branch.

Figure 4(b) shows the peak intensity defined as a sum of $(I_{UFB} + I_{LFB} + I_{const})$ versus $Q$, being almost constant between the high-energy incommensurate scattering. At this stage, no corrections have been made except taking into account a squared magnetic form factor for $Cr^{2+}$ free ions. The $E$-independent scattering $I_{const}$ contributes about half of the above mentioned peak intensity near $T_{sf}$ and it is insensitive to $l$ also. Therefore, the intensity of the FB modes $(I_{UFB} + I_{LFB})$ is independent of $Q$. At
FIG. 5: Contour map around (0, 0, 1) for \( E = 3 \) meV. The intensity around (0, 0, 1.02) is due to the unbalance of the FB mode shown in Fig. 4(a). The inset shows a scan keeping the energy transfer fixed at \( E = 3 \) meV. The triangles point towards the \( \mathbf{Q} \)-positions, where FB excitations were expected according to Ref. \footnote{10}.

The high-\( \mathbf{Q} \) side a transfer of intensity from the upper to the lower FB branch takes place while keeping the total intensity fixed.

Böni \textit{et al.} have already found an asymmetry of the FB mode for the first time \footnote{14} based on an intensity contour map constructed from constant-\( E \) and constant-\( \mathbf{Q} \) scans. They found a blob of scattering at (0, 0, 1.02) near 3 meV. However, the quality of the data did not allow to clearly establish the pronounced (and unexpected) asymmetry in scattering unambiguously \footnote{15}. Our new measurements clearly reproduce the asymmetry between the scattering at (0, 0, 0.98) and (0, 0, 1.02) as shown in Fig. 4 using a different single-\( \mathbf{Q} \) crystal on a different spectrometer with different resolution.

We notice here once more, that it will be difficult to obtain new insight from constant-\( E \) scans as shown in the inset of Fig. 5, unless the incommensurate scattering at \( \mathbf{Q}_c \) can be correctly evaluated and convoluted with the instrumental resolution function. We emphasize that it is the constant-\( \mathbf{Q} \) technique that allows the unambiguous determination of dispersion relations and not the constant-\( E \) technique that leads often to wrong conclusions as proven in the past.

Figures 6 and 7 show the evolution of the inelastic intensity at (0, 0, 1.02) and (0, 0, 0.98) with \( T \), respectively. The magnetic intensity increases much faster than expected on the basis of the thermal population factor, \( \langle n + 1 \rangle = \frac{1}{1 - \exp(-E/k_B T)} \), indicating that the scattering is not due to spin waves \footnote{16}. In particular, a remarkable growth is seen in \( I_{\text{const}} \) as shown with shades. The increase is also not due to conventional critical scattering near \( T_N \) because the measurements were performed far away from \( T_N = 311 \) K, i.e. at reduced temperatures \( t = 1 - (T/T_N) \) of 0.58 and 0.71 for 180 K and 220 K, respectively. We point out that with increasing \( T \) the “missing” peak develops at (0, 0.98) near 3 meV (Fig. 7), i.e. the spectrum becomes more symmetric with increasing \( T \) and approaches the cross section at (0, 0, 1.02) (Fig. 5).

Figure 8 shows (i) once more the asymmetry of the scattering with respect to \( \mathbf{Q}_c \) and (ii) the reduction of unbalance for two peaks at (0, 0, 0.98) with increasing \( T \). These high-resolution measurements (\( \Delta E \leq 0.4 \) meV in FWHM) confirm the results from Fig. 7 (\( \Delta E \leq 1.2 \) meV) that were performed with thermal neutrons. Due to the very different resolution conditions the asymmetry of the scattering is less pronounced with cold neutrons.

**DISCUSSIONS**

We have shown that the FB modes can be measured more precisely by means of constant-\( \mathbf{Q} \) scans than by means of constant-\( E \) scans. As a result, it was possible to clearly identify an asymmetry of the magnetic cross sections with respect to the commensurate position \( \mathbf{Q}_c \). The asymmetry decreases with increasing \( T \). We have proven
by using very different experimental conditions and different spectrometers that the asymmetry is not due to a resolution effect. The asymmetry is also not an artifact of the sample because similar results were obtained using another Cr single crystal and another spectrometer [14]. It might be interesting to investigate the asymmetry of the scattering in other Brillouin zones.

Our results also indicate that the FB modes are not spin-wave modes because the peak intensity is independent of Q (Fig. 4(a)). Moreover, we observe a transfer of intensity between the lower and upper branches of the excitations. Finally, the intensity of the modes increases significantly faster with increasing T than given by ⟨n + 1⟩, which is in agreement with previous works [2, 13]. One may speculate that we have to deal with a thermally activated process. Another possible explanation may be reached by considering mode-mode coupling between the upper and lower FB branches [17].

The present results strongly suggest the existence of magnetic scattering I_{const} that develops around the commensurate Qc position. Its intensity increases also significantly faster than given by ⟨n + 1⟩ and finally dominates the inelastic scattering between Q± in agreement with previous constant-E works [2, 11]. I_{const} should contribute to the significant critical scattering near T_N [2]. In contrast to the FB modes this scattering is polarization independent [4] and exists already in the LSDW phase below the spin-flop transition. The energy-independent feature of I_{const} is also present transverse to Q± but drastically decreases with increasing h and k [12]. We do not know the origin yet. One may speculate that the FB modes as well as the E-independent scattering may be explained by the complicated shape of the Fermi surface that gives rise to many different modes. We mention in particular that already a three-band model gives rise to at least 25 magnetic modes [7].

CONCLUSION

We have reinvestigated the low-energy FB excitations of Cr in the TSDW phase by making extensive use of constant-Q scans. We have observed an asymmetry in the inelastic scattering near T_{sf} that is at variance with the simple centro-symmetric bcc structure of Cr. The asymmetry of energy spectra decreases with raising T. The observed peak intensity of the FB modes satisfies a sum rule with regard to Q. The intensity of the dispersive excitations and the E-independent scattering increase with increasing T much faster than expected according to the temperature factor ⟨n + 1⟩. Therefore, the FB modes are not spin-wave modes. We expect that the present results challenge the development of a theory to explain these results and resolve the puzzle of the magnetic excitations in Cr.

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