New Magnetic Excitations in the Spin-Density-Wave of Chromium

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Low-energy magnetic excitations of chromium have been reinvestigated with a single-\(Q\) crystal using neutron scattering technique. In the transverse spin-density-wave phase a new type of well-defined magnetic excitation is found around \((0,0,1)\) with a weak dispersion perpendicular to the wavevector of the incommensurate structure. The magnetic excitation has an energy gap of \(\omega \approx 4\) meV and at \((0,0,1)\) exactly corresponds to the Fincher mode previously studied only along the incommensurate wavevector.

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The spin-density-wave (SDW) in Cr is one of the most fascinating subjects in condensed matter physics. It has a history of long and continuing research. In spite of the simple body-centered cubic structure with a lattice constant \(a = 2.88\) Å, Cr and its alloys show interesting magnetic behaviors. Below the Neel temperature \(T_N = 311\) K, an incommensurate antiferromagnetic structure develops due to a transverse spin-density-wave (TSDW) with the moments oriented perpendicular to the ordering wavevector \(Q_\perp = (\pm \pi/\alpha)0,1\) (\(\delta \approx 0.048\) at \(T = 100\) K) (see Fig. 1(a)). At \(T_m = 121\) K a spin-flop transition takes place to a longitudinal spin-density-wave (LSDW) phase with the moments along \(Q_\parallel\).

The magnetic cross section in this system also shows a surprisingly rich behavior. The magnetic excitations from these SDW ordered states emerge from the incommensurate positions with high mode-velocities (Fig. 1(a), two cones). In the TSDW phase this metallic antiferromagnet exhibits two types of magnetic fluctuations with the polarization transverse and longitudinal relative to the spin direction. Recently, using polarized neutron scattering in the TSDW phase, it was confirmed that the velocity of the transverse-mode excitations is significantly higher than the velocity of the longitudinal excitations.\(^{[6]}\)

In addition to the incommensurate scattering with large energy scale, Fincher \emph{et al.} observed a resonance like scattering localized at the commensurate position \((0,0,1)\) and at \(\omega = 4\) meV, (Fig. 1(a), open circle) in the TSDW phase.\(^{[6]}\) Later on Burke \emph{et al.} reinvestigated the low energy excitations and concluded that the Fincher-excitation at \((0,0,1)\) was part of dispersion curves for magnetic modes that emanate symmetrically from the \((0,0,1 \pm \delta)\) positions at the incommensurate wavevectors (Fig. 1(a), lines).\(^{[6]}\) Although many neutron scattering experiments have been performed around \((0,0,1)\) and many interesting results were presented, no simple and conclusive explanation was obtained for the origin and details of the

![FIG. 1: Energy dependence of magnetic excitations in the single-\(Q\) TSDW phase of Cr. (a) The cones at the incommensurate positions \((0,0,1 \pm \delta)\) indicate the high-velocity spin excitations. The solid lines represent the proposed dispersion of the FB mode, which cross at the commensurate position \((0,0,1)\) at \(\approx 4\) meV. (b) The data points for the measurements transverse to \(\mathbf{Q}_\perp\) indicate excitations with a gapped dispersion. The inset defines the scans in reciprocal space. Most measurements have been performed around the \((0,0,1)\) Bragg point.](image-url)
and iii) the dispersion below 4 meV is absent, i.e. the FB-mode has an energy gap of 4 meV. However, there still exists substantial disagreement between different experiments, which precludes a full understanding of the magnetic excitations of Cr.

Similar to the experimental side there exist contrasting discussions between theories even in the ground state. For example, although the incommensurate ordering can be explained by the nesting properties of the electron and hole Fermi-surfaces, a recent density-functional investigation predicts a commensurate structure. Concerning the variety of magnetic excitations Fishman and Liu succeeded in calculating the incommensurate excitations and assigning the longitudinal modes as being phonon modes. In addition, they predicted a large number of possible interband transitions. However, since the accuracy of present-day band-calculations does not allow to calculate the low energy spectrum in Cr with high-precision, there is still no acceptable model to explain the FB-excitations.

In this paper, we report a new type of magnetic excitations in the SDW state of Cr. The low energy magnetic excitations of Cr were explored using a large single crystal with a single-Q structure. The original main target was to study the magnetic cross section at the so-called "silent" satellites first investigated by Sternlieb et al. We, therefore, have taken the data near the incommensurate positions (±δ, 0, 1). Quite surprisingly, the well-defined gapped energy spectrum at (±δ, 0, 1) was also observed with a weak dispersion with Q along the Fincher excitation at (0, 0, 1) as shown in Fig. 2(b). Therefore, the Fincher mode has a clear dispersion perpendicular to the incommensurate wavevectors. This new observation of a gaped excitation provides new restrictions on the origin of the FB-mode and definitely buries all previous interpretations.

The inelastic neutron scattering was performed using a large cylindrical single crystal of Cr from Johnson-Matthey Co. with a diameter of 10 mm, a length of 50 mm along [0, 1, 0] direction (V ≈ 4 cm³) and a mosaic n ≈ 40° on the triple-axis spectrometer TOPAN at JRR-3M in Tokai, Japan. In order to produce a single-Q sample the crystal was cooled through T_N in a field of 14 T. The field work was kindly accomplished in cooperation with the High Field Laboratory for Superconducting Materials, Tohoku University. The crystal was aligned with the [1, 0, 0] and the [0, 0, 1] crystallographic directions in the scattering plane. The [0, 0, 1] direction was selected to be parallel to Q±. Note that due to the cylindrical shape of the single crystal along [0, 1, 0] nonmagnetic background could be reduced by narrowing the horizontal beam width. The population of single-Q domain was estimated to be more than 99% from the intensity ratio of the magnetic satellites around the (0, 0, 1) and (1, 0, 0). The final energy of TOPAN was fixed at 14.7 meV. Two types of horizontal collimation sequence were utilized, Blank(60°)-30°-60°-Blank(100°) and 30°-30°-10°-Blank(100°) from before the monochromator to after the analyzer. The energy resolution of each collimation is evaluated to be 1.4 and 0.8 meV in FWHM, respectively. Higher order neutrons were removed by means of a pyrolytic graphite filter. Furthermore, in order to reduce the high-energy neutron background a sapphire single crystalline filter was inserted in between the first and second Soller collimators.

Typical scans for scattering vectors Q along (scan A) and perpendicular (scan B) to the incommensurate ordering vector Q± in the TSDW (T = 140 K) and LSDW (T = 100 K) phases. The Fincher-mode is observed by the scan A only in the TSDW phase (open circles in (a)). The weak commensurate scattering intensity in the LSDW phase of (a) does not signify any excitations (see text). (Ref. Fincher et al. [6]).

**FIG. 2:** The scans for constant ω = 4 meV with scattering vectors Q (a) along (scan A in the inset) and (b) perpendicular (scan B in the inset) to the incommensurate ordering vector Q± in the TSDW (T = 140 K) and LSDW (T = 100 K) phases. The Fincher-mode is observed by the scan A only in the TSDW phase (open circles in (a)). The weak commensurate scattering intensity in the LSDW phase of (a) does not signify any excitations (see text). (Ref. Fincher et al. [6]).
LSDW phase ($T = 100\, \text{K}$) is not due to the Fincher-mode as the intensity at $(0, 0, 1)$ exhibits no appreciable energy dependence (Figs. 3(a) and 3(a)). The Fincher mode is easily observed on top of the energy independent magnetic intensity (broken line in Fig. 3(a)). On the other hand, in the scan B no remarkable difference is seen between the two phases. The Fincher-mode is therefore not observed in this scan. On this aspect many detailed discussions have already been made. Note that in the previous scans along $Q_\pm$ the weak signal from the FB-mode is difficult to detect due to the steep "background" from the incommensurate peaks.

In order to get a clue about the origin of the FB-mode we explored the $Q$-dependence of the Fincher excitation perpendicular to $Q_\pm$ (scan (b) in the inset of Fig. 2). Some typical scans measured at $T = 140\, \text{K}$ and $T = 100\, \text{K}$ are shown in Fig. 2. A well-defined peak is observed in the TSDW phase at $T = 140\, \text{K}$. Quite surprisingly, the peak energy clearly moves to larger $\omega$ with increasing transverse momentum (see Fig. 2(b)) accompanied by a substantial decrease in the intensity. The peak-width in Fig. 2 is broader than the instrumental resolution width. It is noted that a well-defined signal was obtained due to the focusing effect of the instrumental resolution.

Because of this interesting observation we decided to map out the magnetic scattering in more detail and performed constant-$Q$ scans to construct contours. As shown in Fig. 3(a), we can get a clear excitation spectrum with high-resolution, which confirms the single-peaked Fincher excitation. However, we tuned the spectrometer with the medium collimator sequence so that we could get a reasonable statistics for the contours. We note that the signal-to-noise ratios in Figs. 3(a) and 3(a) do not change so much even in the different instrumental resolutions. The intensity of the new mode as well as the Fincher-mode were evaluated by subtracting the nearly constant intensity (broken lines in Fig. 3) as described before. The resulting intensity contours are shown in Fig. 3(b). One can follow the dispersion relation out to $\omega = 0.1$, i.e. about twice as far as the incommensurability $\delta$ of the spin-density wave.

Our results establish a new magnon branch centered around the Fincher-mode at $(0, 0, 1)$ and shows a weak dispersion along the $h$ and $k$ direction, i.e. perpendicular to the magnetic ordering vector $Q_\pm$. In contrast to previous scans along the $l$ direction the data is rather clean because there is no strong incommensurate scattering near the "silent" positions ($\pm \zeta, 0, 1$). The new mode has the following important properties: i) It has an energy gap of 4 meV and can only be observed in a relatively narrow $Q$ and $\omega$ range. ii) The new mode exists only in the TSDW phase demonstrating that it is only allowed due to the transverse orientation of the magnetic moments with respect to $Q_\pm$, i.e. the spin-flop transition opens a new degree of freedom for excitations. iii) The mode has the same longitudinal polarization as the Fincher-mode implying once more that it is intimately connected with the ordering of the spin-density-wave. It is clear these results are not compatible with any interpretations for the FB-mode given in the literature so far.

The non-existence of the mode in the LSDW-phase and the gap in the TSDW-phase bear some similarity to optical phonon modes in insulators that show different dispersions depending on their polarization being transverse or longitudinal with respect to the direction of propagation. One may speculate that it is possible to excite domain walls (or stripes) in the TSDW phase that require a nucleation energy of about 4 meV and propagate perpendicular to $Q_\pm$ thus causing a dispersion. This process may be energetically less favorable in the LSDW-phase.

We point out that the new mode has also some intriguing similarities to the resonance-like peaks in the dynamical magnetic susceptibility observed for the sev-
FIG. 4: (a) High-resolution energy spectra at (0, 0, 1). The Fincher excitation in the TSDW state \((T = 140 \text{ K})\) shows a single peak. (b) Contour-map of the magnetic scattering due to the Fincher excitations, measured with the medium resolution. It shows a gapped dispersion that extends with increasing \(\zeta\) towards 8 meV. The energy-independent intensities are subtracted.

General systems with strongly correlated electrons; high-\(T_c\) cuprates such as \(\text{YBa}_2\text{Cu}_3\text{O}_{7-y}\) \([\text{17}]\) that orders also in an incommensurate structure, and the geometrically frustrated \(\text{ZnCr}_2\text{O}_4\) \([\text{18}]\). We hope that our results encourage new efforts to understand the antiferromagnetic state in Cr, in particular the interplay between this new mode and the enhancement of the longitudinal incommensurate scattering and the scattering at the silent positions, which will also elucidate the common aspect of magnetic excitations in strongly correlated electron systems.

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