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Reinvestigation of 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-$\boldsymbol{Q}_{\pm}$ crystal using neutron scattering techniques. In the transverse spin-density-wave phase a well-defined magnetic excitation is found around (0,0,1) with a weak dispersion *perpendicular* to the wave vector of the incommensurate structure. The magnetic excitation has an energy gap of $\omega \simeq 4$ meV at (0,0,1), which exactly corresponds to the Fincher excitation previously studied only *along* the incommensurate wave vector.

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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.[1] 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.[2,3] Below the Néel 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 wave vector $\boldsymbol{Q}_{\pm}=(2\pi/a)(0,0,1\pm\delta)$ where $\delta \simeq 0.048$ at $T=100$ K (the inset of Fig. 1, open circles). At $T_{\mathrm{SF}}=121$ K a spin-flop transition takes place to a longitudinal spin-density-wave (LSDW) phase with the moments along $\boldsymbol{Q}_{\pm}$.

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.[4]

In addition to the incommensurate scattering with large energy scale, Fincher *et al*. observed a resonancelike scattering (Fincher excitation) localized at the commensurate position (0,0,1) and at $\omega=4$ meV, [Fig. 1(a), solid circle] in the TSDW phase.[5,6] Later on Burke *et al*. reinvestigated the low-energy excitations and concluded that the Fincher excitation at (0,0,1) was part of dispersion lines for magnetic modes [Fincher-Burke (FB) mode] that emanate symmetrically from the $(0,0,1\pm\delta)$ positions at the incommensurate wave vectors [Fig. 1(a), lines].[7] Furthermore, the neutron scattering measurement with full polarization analysis observed well-defined peaks with $\omega=6$ meV at $(0,0,1\pm0.02)$ consistent with the postulated FB mode.[8]

Although many other neutron-scattering experiments have been also performed around (0,0,1) (Refs. 9–14) and many interesting results were presented, no simple and conclusive explanation was obtained for the origin and details of the FB mode. Among them the most recent polarized-beam and high-resolution measurements provide several constraints for the FB mode: (i) the mode has longitudinal polarization,[4,9] (ii) the symmetric branches for $Q<Q_-$ and $Q>Q_+$ do not exist,[10] and (iii) the well-defined dispersion below 4 meV is absent, i.e., the FB mode has an energy gap of 4 meV.[14] However, there still exists substantial disagreement between different experiments, which precludes a full understanding of the magnetic excitations of chromium.

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,[15] a recent density-functional investigation predicts a commensurate structure.[16] Concerning the variety of magnetic excitations Fishman and Liu[17] succeeded in calculating the incommensurate excitations and assigning the longitudinal modes as being phason modes. In addition, they predicted a large number of possible interband transitions. However, since the accuracy of present-day band-calculations does not allow calculation of the low energy spectrum in Cr with high precision, there is still no acceptable model to explain the Fincher excitation as well as the FB mode.

In this paper, we report on a newly found magnetic excitation that appears in the TSDW state of Cr. (We hereafter call it the ‘‘new mode.’’) The low-energy magnetic excitations of Cr were explored using a large single crystal with a single-$\boldsymbol{Q}_{\pm}$ structure. The new mode is observable around the Fincher excitation, but qualitatively different with the previ-

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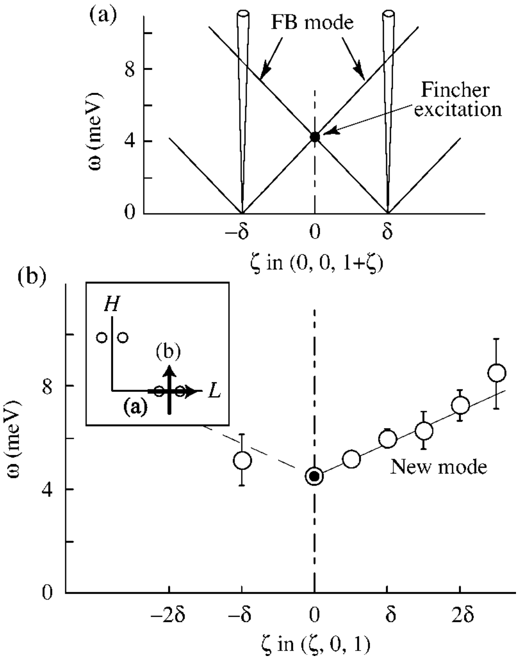

FIG. 1. Energy dependence of magnetic excitations in the single-$\boldsymbol{Q}_{\pm}$ TSDW phase of Cr. (a) The cones at the incommensurate positions $(0,0,1\pm\delta)$ indicate the high-velocity spin excitations. The lines represent the proposed dispersion of the FB mode, which cross at the commensurate position at $\simeq 4$ meV (Fincher excitation, solid circle). (b) The data points for the measurements transverse to $\boldsymbol{Q}_{\pm}$ 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. See the detailed reciprocal lattice of bcc Cr and the polarization direction of the SDW states depicted in Refs. 4,14.

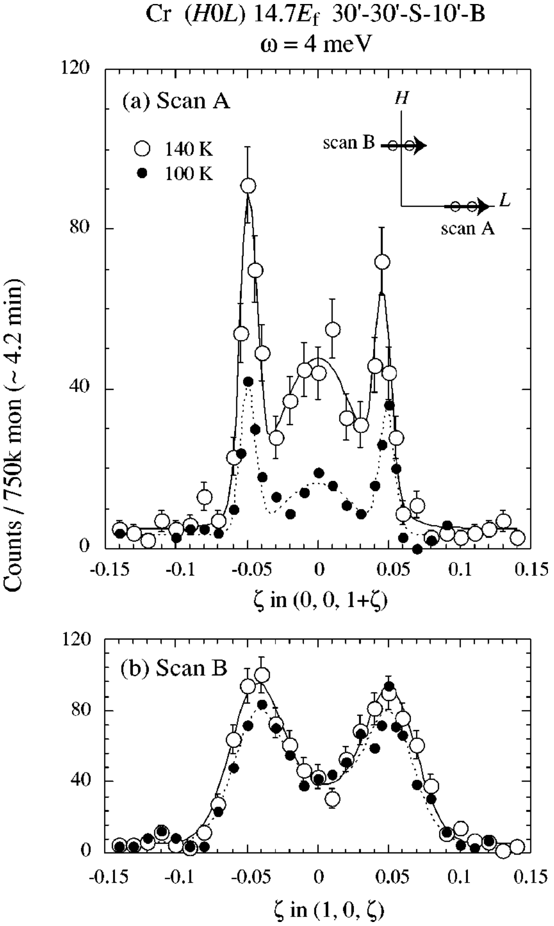

FIG. 2. The scans for constant $\omega = 4$ meV with scattering vectors $\boldsymbol{Q}$ (a) along (scan A in the inset) and (b) perpendicular (scan B in the inset) to the incommensurate ordering vector $\boldsymbol{Q}_{\pm}$ in the TSDW ($T = 140$ K) and LSDW ($T = 100$ K) phases. The Fincher excitation 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) is regarded not as the Fincher exciattion (see the text.)

ously published FB mode. First, it forms a clear dispersion *perpendicular* to $\boldsymbol{Q}_{\pm}$. [Figures 1(b) and 4]. Secondly, the dispersion relation extends out to 2δ from (0,0,1). Thirdly, it indeed shows a gapped dispersion. The new mode is observed only in a narrow $(\boldsymbol{Q},\omega)$ range and it disappears in the LSDW state. These facts are neither theoretically predicted nor experimentally observed in the previous studies. Therefore, it is required to reconstruct the picture of the low-energy magnetic excitation for Cr.

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 ($V \simeq 4$ cm^3), and a mosaic $\eta \simeq 40'$ on the triple-axis spectrometer TOPAN at JRR-3M in Tokai, Japan. In order to produce a single-$\boldsymbol{Q}_{\pm}$ 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 $\boldsymbol{Q}_{\pm}$. 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 a single-$\boldsymbol{Q}_{\pm}$ 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 full width at half maximum (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 $\boldsymbol{Q}$ along (scan A) and perpendicular (scan B) to the incommensurate vector $\boldsymbol{Q}_{\pm}$ are shown in Figs. 2(a) and 2(b), respectively. As seen in Fig. 2(a), the scattering in the TSDW phase ($T = 140$ K) is composed of the Fincher excitation seen at around (0,0,1) and incommensurate peaks at $(0,0,1\pm\delta)$. In this scan there exists a substantial difference in the intensities between the TSDW and LSDW phases. The broad scattering centered at

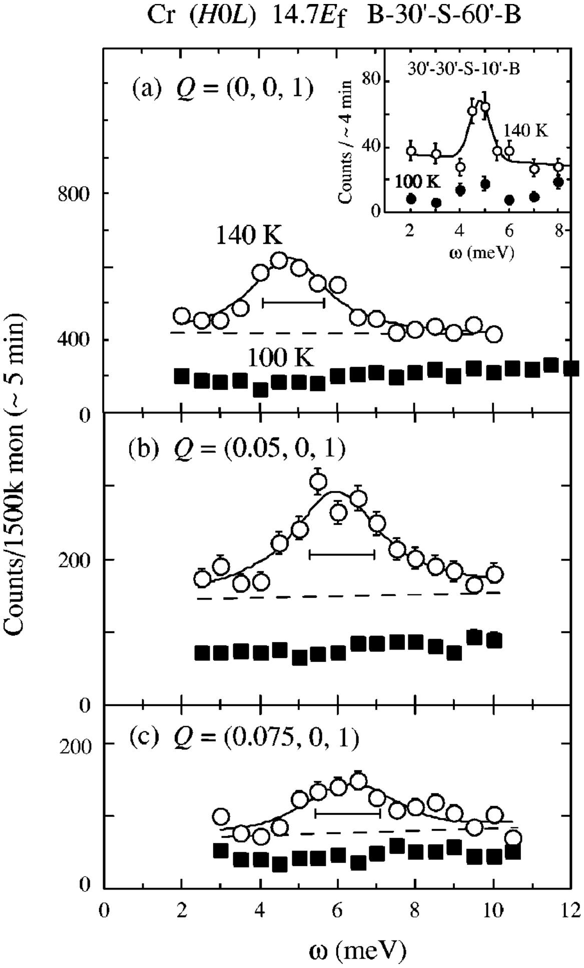

FIG. 3. Constant-$\mathbf{Q}$ scans along the $[\zeta,0,0]$ direction around (0,0,1). In the LSDW phase the scattering is small (filled squares) and independent of ω. The scattering in the TSDW phase (open circles) clearly shows a peak that moves to higher ω with increasing ζ. Each horizontal bar below the peak represents the energy resolution. The inset of (a) shows the high-resolution energy spectra at (0,0,1), displaying the sharp Fincher excitation.

(0,0,1) in the LSDW phase ($T=100$ K) is not due to the Fincher excitation as the intensity at (0,0,1) exhibits no appreciable energy dependence [Fig. 3(a)]. The Fincher excitation 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 excitation is therefore not observed in this scan. On this aspect many detailed discussions have already been made. Note that in the previous scans along $\mathbf{Q}_{\pm}$ 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 $\mathbf{Q}$ dependence of the Fincher excitation perpendicular to $\mathbf{Q}_{\pm}$ [scan (b) in the inset of Fig. 1]. Some typical scans measured at $T=140$ K and $T=100$ K are shown in Fig. 3. A well-defined peak is observed in the TSDW phase at $T=140$ K. Quite surprisingly, the peak energy clearly moves to larger ω with increasing transverse momentum [see Fig. 1(b)] accompanied by a substantial decrease in the intensity. The peak width in Fig. 3 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.

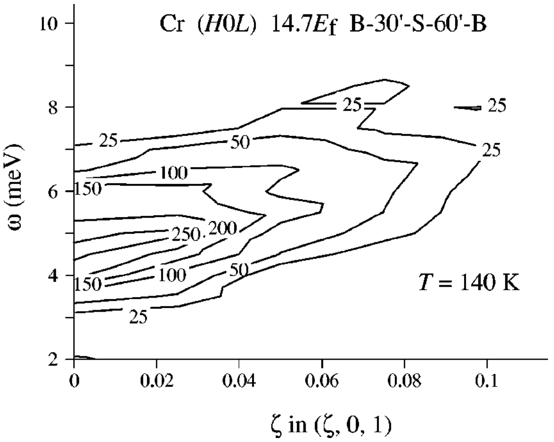

FIG. 4. Contour map of the magnetic scattering due to the Fincher excitation and the new mode, measured in the direction perpendicular to $\mathbf{Q}_{\pm}$. It shows a gapped dispersion that extends with increasing ζ towards 8 meV. The energy-independent intensities are subtracted.

Because of this interesting observation we decided to map out the magnetic scattering in more detail and performed constant-$\mathbf{Q}$ scans to construct contours. As shown in the inset of Fig. 3(a), a well-defined excitation peak is observable using tight beam collimation, which confirms the single-peaked Fincher excitation. However, we tuned the spectrometer with the medium collimator sequence so as to obtain resonable statistics for the contours. We note that the signal-to-noise ratios in Fig. 3(a) and the inset do not change so much even in the different instrumental resolutions. The intensity of the new mode as well as the Fincher excitation was evaluated by subtracting the nearly constant intensity (broken lines in Fig. 3) as described before. (In the future, we will examine the detail of the energy-independent scattering.) The resulting intensity contours are shown in Fig. 4. One can follow the dispersion relation out to $\zeta \simeq 0.1$, i.e., about twice as far as the incommensurability δ of the spin-density wave.

Our results establish a new magnon branch centered around the Fincher excitation at (0,0,1) and shows a weak dispersion along the h direction, i.e., perpendicular to the magnetic ordering vector $\mathbf{Q}_{\pm}$. In contrast to previous scans along the l direction, the data is rather clean because there is no strong incommensurate scattering near the "silent" satellite positions of $(\pm\delta,0,1)$.[12] 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 $\mathbf{Q}$ and ω 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 $\mathbf{Q}_{\pm}$, 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 excitation implying once more that it is intimately connected with the ordering of the spin-density wave. It is clear that these results are not compatible with any interpretations for the FB mode given in the literature so far.

The nonexistence of the new 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 perpendicularly to $\boldsymbol{Q}_{\pm}$ thus causing a dispersion. This process may be energetically less favorable in the LSDW phase.

We point out that the localized new mode has also some intriguing similarities to the resonance like peaks in the dynamical magnetic susceptibility observed for the several systems with strongly correlated electrons; high-T_c cuprates such as $YBa_2Cu_3O_{7-y}$ (Ref. 18) that orders also in an incommensurate structure, and the geometrically frustrated $ZnCr_2O_4$.[19] Finally, 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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