Magnetic Excitations in the Incommensurate Phases of Chromium Metal

S. K. Burke,[a] W. G. Stirling, and K. R. A. Ziebeck
Institut Laue-Langevin, F-38042 Grenoble, France

and

J. G. Booth
Department of Pure and Applied Physics, University of Salford, Salford, United Kingdom

(Received 23 May 1983)

The excitation spectrum of the incommensurate spin-density wave in chromium metal has been investigated by neutron scattering. It is found that the high-velocity mode originating from the magnetic satellite positions is not a simple spin wave but at low frequency is a longitudinal excitation. Additional excitation branches, which cross at the (100) position at a frequency corresponding to the "commensurate" feature seen in previous studies, were also observed.

PACS numbers: 75.30.Ds, 75.50.Ee

The unusual magnetic properties of chromium metal, a sinusoidally modulated antiferromagnet, have been the subject of intense theoretical and experimental study dating back to the original proposal of antiferromagnetism by Néel in 1932. It is now widely accepted that the antiferromagnetic state results from certain geometrical features of the paramagnetic Fermi surface which favor a collective instability of the *sd* conduction electrons to form, below the Néel temperature ($T_N = 311$ K), a spin density wave (SDW) with a wave vector $\vec{Q}$ which is incommensurate with the lattice.[1] Theoretical descriptions of Cr based on these concepts successfully account for the ordering wave vector, ordering temperature, and ordered moment in both pure Cr and its alloys.[2] Despite these achievements, relatively little progress has been made in understanding the elementary magnetic excitations of the SDW. Present theories[3-5] are conflicting and do not reproduce even the most basic features observed by experiment.[6-9] In view of this outstanding fundamental problem we set out to examine the magnetic excitation spectrum of pure Cr in more detail.

The neutron inelastic-scattering measurements were performed with the IN8 and IN12 triple-axis spectrometers at the Institut Laue-Langevin, Grenoble. A curved Cu(111) monochromator and pyrolytic graphite (PG) (002) analyzer were used for the IN8 experiments. The collimations between monochromator and sample, sample and analyzer, and analyzer and detector were all 40′. Data were collected with either the final neutron wave vector (K_F) fixed at 2.66 and 3.5 Å^{-1} or the incident wave vector (K_I) fixed at 2.66 Å^{-1}. Measurements at low frequency transfer (ν) were made with IN12. Here, a curved PG(002) monochromator, collimations of 30′, and a PG(002) analyzer were used; K_I was fixed at 1.55 Å^{-1}. PG or beryllium filters were employed to reduce higher-order contamination.

The cylindrical single crystal of chromium was grown from high-purity iodide-process material: The cylinder axis was [010], the volume 1.5 cm^3, and mosaic spread 20′. Measurements were made in the (010) scattering plane. Prior to the inelastic-scattering experiments, the crystal was cooled through T_N in an applied magnetic field of 15 T parallel to [100] to produce a 99% single-$\vec{Q}$ state as determined from the relative intensities of the $(1\pm\delta,0,0)$ and $(0,0,1\pm\delta)$ satellites (i.e., an unbalanced Q-domain population in which the volume of [100] Q domains was about 99% of the total).

The magnetic structure of bcc Cr is based on the sinusoidal spin polarization of the $3d$ electrons on the two simple-cubic sublattices. The modulation wavelength is typically 20 to 28 unit cells and $\vec{Q}$ is slightly less than a (100) reciprocal lattice vector, $|\vec{Q}| = (1-\delta)2\pi/a$ where $\delta \approx 0.05$.[10] Below the "spin-flip" temperature ($T_{SF} = 121$ K), in the longitudinal SDW (LSDW) phase, the magnetic moments are parallel to $\vec{Q}$, whereas at higher temperature ($T_{SF} < T < T_N$), in the transverse SDW (TSDW) phase, the magnetic moment and $\vec{Q}$ lie along orthogonal cube axes. The (010) reciprocal lattice plane of single-Q Cr is shown in Fig. 1(c).

Attention was focused initially on the stiff magnetic excitations which emerge from the magnetic satellite positions. In agreement with previous work,[6,8] we observe these modes to move slightly inwards toward the (100) position, with a velocity of 110 ± 10 THz Å, as the frequency is increased.

The polarization of these modes (i.e., the direction of spin fluctuations) was determined by measuring the intensity of the excitations around the satellite positions $(1\pm\delta,0,0)$, where the scattering vector $(\vec{K})$ is parallel to $\vec{Q}$, and $(\pm\delta,0,1)$ where $\vec{K}$ is almost perpendicular to $\vec{Q}$. Only the spin fluctuations transverse to $\vec{K}$ contribute to the magnetic scattering cross section. In the LSDW phase, as shown in Fig. 1(a), the intensity of the scattering at $(1-\delta,0,0)$ tends to zero at low frequency whereas the scattering at comparable frequencies at $(\delta,0,1)$ remains large. (The mode at 1.9 THz will be discussed later.) Thus, at low frequencies, the spin fluctuations are parallel to $\vec{Q}$ and hence to the ordered moment. This observation is important as it shows that the high-velocity mode in the LSDW phase, at low frequency, is one of the very few examples of a *longitudinal* magnetic excitation.[11] In the TSDW phase, the integrated intensities of the modes at $(1-\delta,0,0)$ and $(\delta,0,1)$ were determined under

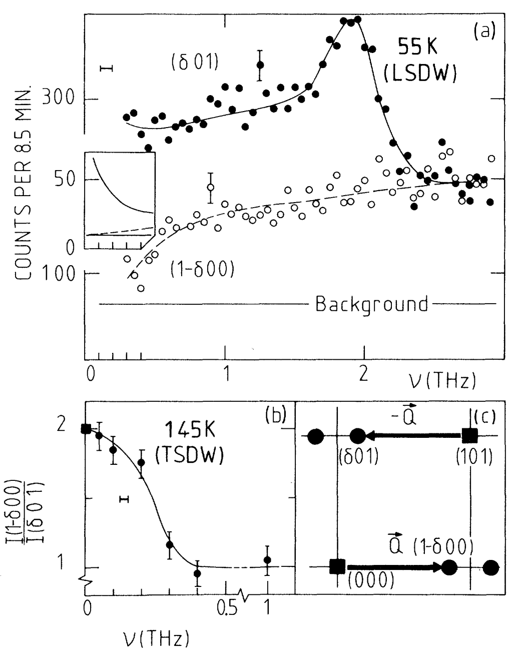

FIG. 1. (a) LSDW phase. Constant-Q scans at the $(\delta, 0,1)$ and $(1-\delta,0,0)$ satellite positions at $T=55$ K, $K_F = 2.66$ Å^{-1} (IN8). Inset: Results at low frequency, $K_I = 1.55$ Å^{-1} (IN12) on the same frequency scale. The increase in inentsity at very low frequencies at $(\delta,0,1)$ arises from the magnetic satellite. The curves are to guide the eye and the background level is shown.
(b) TSDW phase. Ratio of corrected integrated intensities of the excitations at $(1-\delta,0,0)$ and $(\delta,0,1)$. At the former of these wave vectors, the scan direction was [001]; at the latter, the scan direction was [100].
(c) (010) reciprocal lattice plane of single-$\vec{Q}$ Cr. Circles and squares are respectively magnetic and nuclear Bragg positions.

similar conditions of resolution in constant-ν scans. The ratio of these intensities, corrected for the difference in magnetic form factor between the two positions, is shown as a function of frequency in Fig. 1(b). This ratio tends to a value of 2 at low frequency indicating that the spin fluctuations are perpendicular to $\vec{Q}$. As two types of transversely polarized magnetic domains are present in this phase, it is not possible to determine unambiguously whether these fluctuations are also parallel to the ordered moment. As the frequency is increased, in both phases, the polarization changes continuously until at sufficiently high frequency the spin fluctuations become isotropic and the intensities of the $(1\pm\delta, 0,0)$ and $(\pm\delta,0,1)$ modes are equal.

Although a number of theoretical studies have predicted the existence of longitudinal spin waves in itinerant antiferromagnets,[5,12] they do not account for the present observation that longitudinal modes dominate the low-frequency response of LSDW Cr, as gapless transverse excitations are always predicted to accompany the longitudinal excitations. These simplified models ignore two important points. Firstly, longitudinal excitations in an incommensurate spin system may arise not only from fluctuations in the *amplitude* of the moment but also via fluctuations in the *phase* of the modulation, and secondly, magnetic anisotropy is neglected. The latter introduces a gap in the transverse excitation spectrum. Of the two longitudinal modes, the phase mode may be loosely identified with the Goldstone mode of the SDW. We suggest that it is this longitudinal phase

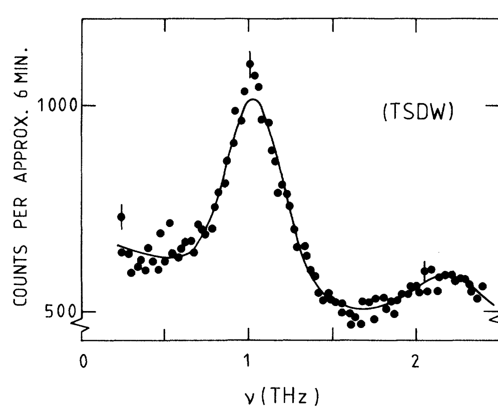

FIG. 2. Constant-Q scan at $(1,0,0)$ at $T=230$ K, $K_F = 2.66$ Å^{-1} (IN8). As well as the "commensurate feature" at about 1 THz, note the higher-energy peak at about 2.2 THz which was also seen in the work of Ref. 7.

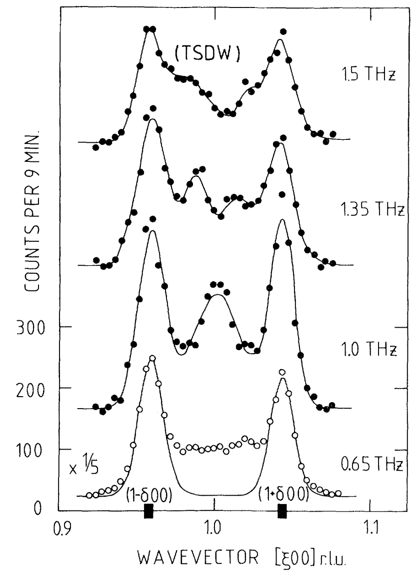

FIG. 3. Constant-ν scans through the (100) position at $T=230$ K. The scan direction is parallel to [100]. Solid circles, $K_I=2.66$ Å^{-1}; open circles, $K_F=2.66$ Å^{-1}. The solid curves are fits to the data using Gaussian profiles.

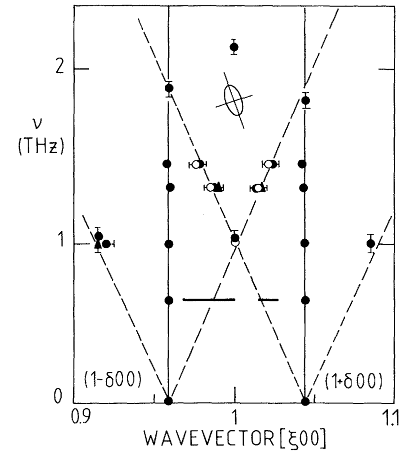

FIG. 4. TSDW phase, dispersion relation. The high-velocity excitation (solid lines) has been observed in previous studies. The additional branches of excitations found in the present work are shown by the dashed lines. The projection of the resolution ellipsoid for $K_F=2.66$ Å^{-1} is also shown. Solid circles, $K_F=2.66$ Å^{-1}; open circles, $K_I=2.66$ Å^{-1}; triangles, $K_F=3.5$ Å^{-1}.

excitation which is responsible for the observed low-frequency response of Cr. The second main feature of the (magnetic) excitation spectrum is the "commensurate" mode reported to occur at a frequency of ~ 1 THz at the commensurate {100} positions in the TSDW phase.[6-7] Figure 2 shows this commensurate mode as observed in the present experiment. The inelastic response in the vicinity of this peculiar feature has more structure than hitherto expected. This is demonstrated by the results of constant-ν scans parallel to [100] through the (100) position, Fig. 3. The three-peak structure at 1 THz evolves into four peaks in scans at 1.35 and 1.5 THz as the commensurate excitation splits into two distinct modes moving outwards towards the $(1\pm\delta,0,0)$ spin-wave peaks.

The dispersion relation for these excitations is given in Fig. 4. This represents a compilation of results obtained under varying spectrometer conditions, as indicated. To within the fitting error the excitations lie at the same positions independent of the instrument configuration. The most striking feature of the dispersion curves is that the excitations apparently originating from the commensurate excitation are consistent also with linear dispersion from the satellite positions. Indeed, a constant-ν scan at 0.65 THz shows a plateau of intensity between the two spin-wave modes consistent with excitations entering the commensurate mode and further, excitations are observed propagating in the opposite direction with the same velocity ($\xi\sim 0.9$ and 1.1). Hence the commensurate mode, rather than being an isolated, discrete excitation of the incommensurate SDW,[6] is a particular point of high intensity arising from the crossover of two branches of propagating excitations.

The velocity of these excitations, 10.5 ± 0.5 THz Å, is identical to that of the [ξ00] longitudinal acoustic (LA) phonon.[13] This suggests that they may be due to magnetovibrational scattering (scattering which is elastic in the spin system but inelastic in the phonon system[14]) and thus they represent no more than the LA phonon viewed through magnetic interactions with the neutron. This would account for the disappearance of the commensurate mode at (100) in the LSDW phase and also the temperature dependence of the mode frequency.[6] We note that these modes are still observed around (001) in the LSDW phase as was shown in Fig. 1(a). Our explanation does not, however, explain the marked temperature dependence of the intensity,[7] which may imply some coupling between the spin and phonon systems. It is interesting to note that magnetostriction and thermal expansion measurements in Cr[15] have been interpreted by means of a model involving the thermal activation of "polarization domains." Further measurements are required to clarify these points.

We are grateful to R. Pynn, R. Currat, K. A.

McEwen, D. E. Okpalugo, and S. K. Sinha for helpful discussions. The Cr crystal was grown by Metal Crystals Ltd., Cambridge, United Kingdom, and the field-cooling procedure was carried out at Service National des Champs Intenses, Grenoble. This work was supported in part by the Science and Engineering Research Council, United Kingdom.

[(a)]Present address: Materials Division, Aeronautical Research Laboratories, Melbourne, Australia.

[1]W. M. Lomer, Proc. Phys. Soc. London 86, 489 (1962).

[2]See for example E. W. Fenton and C. R. Leavens, J. Phys. F 10, 1853 (1980).

[3]T. Wolfram and S. Ellialtioglu, Phys. Rev. Lett. 44, 1295 (1980).

[4]S. H. Liu, J. Magn. Magn. Mater. 22, 93 (1980).

[5]H. Sato and K. Maki, Int. J. Magn. 6, 183 (1974).

[6]C. R. Fincher, Jr., G. Shirane, and S. A. Werner, Phys. Rev. Lett. 43, 1441 (1979), and Phys. Rev. B 24, 1312 (1981).

[7]S. A. Werner, G. Shirane, C. R. Fincher, and B. H. Grier, in *Neutron-Scattering—1981*, edited by John Faber, Jr., AIP Conference Proceedings No. 89 (American Institute of Physics, New York, 1982), p. 269.

[8]J. G. Booth and K. R. A. Ziebeck, J. Phys. F 9, 2424 (1979).

[9]C. Windsor, Physica (Utrecht) 91B, 119 (1978).

[10]S. A. Werner, A. Arrott, and H. Kendrick, Phys. Rev. 155, 528 (1967).

[11]Longitudinal excitations, in the form of propagating crystal field modes, are found in many rare-earth metals and compounds and are well understood. In the present case, that of a $3d$ metal, we know of no other well established examples.

[12]J. B. Sokoloff, Phys. Rev. 185, 770, 783 (1969), and Phys. Rev. 187, 584 (1969).

[13]W. M. Shaw and L. D. Muhlestein, Phys. Rev. B 4, 969 (1971).

[14]W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Clarendon, Oxford, 1971), p. 233.

[15]M. O. Steinitz, L. H. Schwartz, J. A. Marcus, E. Fawcett, and W. A. Reed, Phys. Rev. Lett. 23, 979 (1969).