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EXCITATIONS OF THE SPIN-DENSITY WAVE IN PURE CHROMIUM

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ABSTRACT

This paper summarizes our recent investigations of the magnetic excitations of the spin density wave (SDW) in pure Cr in both the low temperature longitudinally polarized phase (T < 122K) and in the higher temperature transversely polarized phase (122K < T < 312K). In both phases we observe spin wave modes of very high velocity originating from the incommensurate Bragg points. In the transversely polarized SDW phase new additional excitations are observed, centered in reciprocal space at the (1,0,0) commensurate point. These excitations are not affected by a magnetic field. Inelastic scattering in the paramagnetic phase above the Neel point (312K) is observed in a reasonably well localized region of reciprocal space near (1,0,0) indicating that there are spin-spin correlations extending over many bcc unit cells and persisting to temperatures at least as high as 1.7 T_N.

DISCLAIMER

INTRODUCTION

The itinerant magnetism in chromium metal is due to an Overhauser spin density wave (SDW).[1] As first pointed out by Lomer[2] this unique antiferromagnetic state occurs in chromium because of the nearly perfect nesting of portions of the jack-shaped electron Fermi surface surrounding the Γ-point (0,0,0) in reciprocal space with the flat portions of the octahedral-shaped hole surface surrounding the H-point (1,0,0). The discovery of the incommensurate nature of the antiferromagnetism in Cr was made 22 years ago by Corliss, Hastings and Weiss,[3] when they identified by neutron diffraction the magnetic satellite structure in which Bragg peaks are found at neutron scattering vectors

$$\vec{Q}_N = \vec{G} \pm \vec{Q}. \tag{1}$$

Here $\vec{Q}$ ($\approx .95 \frac{2\pi}{a}$) is the wave vector of the SDW and $\vec{G}$ is any reciprocal lattice vector of the body centered cubic lattice of Cr. The resulting (100) reciprocal lattice plane is shown in Fig. 1. There is now a very large body of literature on investigations of the magnetic properties of this extremely interesting metal.

There are three magnetic phase of Cr. At temperatures below the spin-flip temperature (T_{sf} = 122K) the SDW is longitudinally polarized. Above T_{sf} the SDW is transversely polarized up to the first order transition at the Neel point (T_N = 312K)[4]. As a result of the cubic symmetry of paramagnetic chromium above T_N, three types of

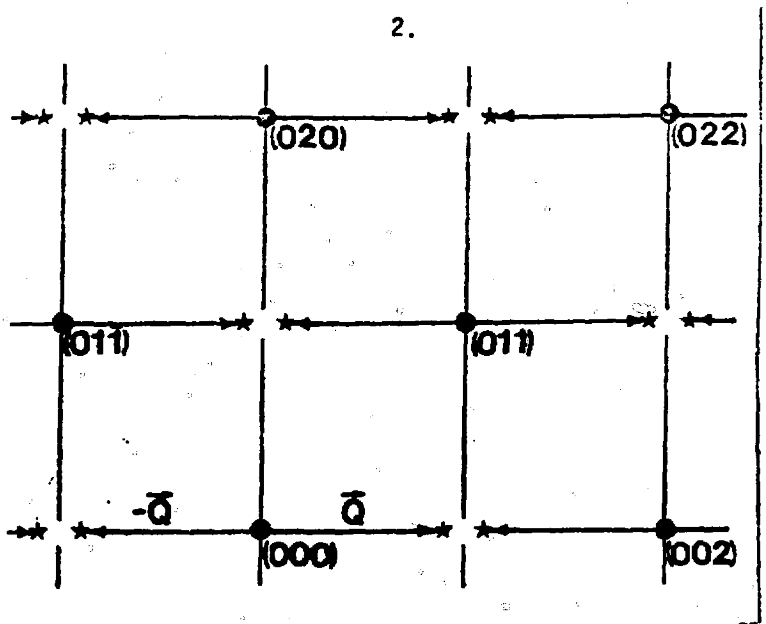

Fig. 1. The (100) reciprocal lattice plane of antiferromagnetic Cr metal. The solid circles are nuclear Bragg reflections. The stars are magnetic Bragg points.

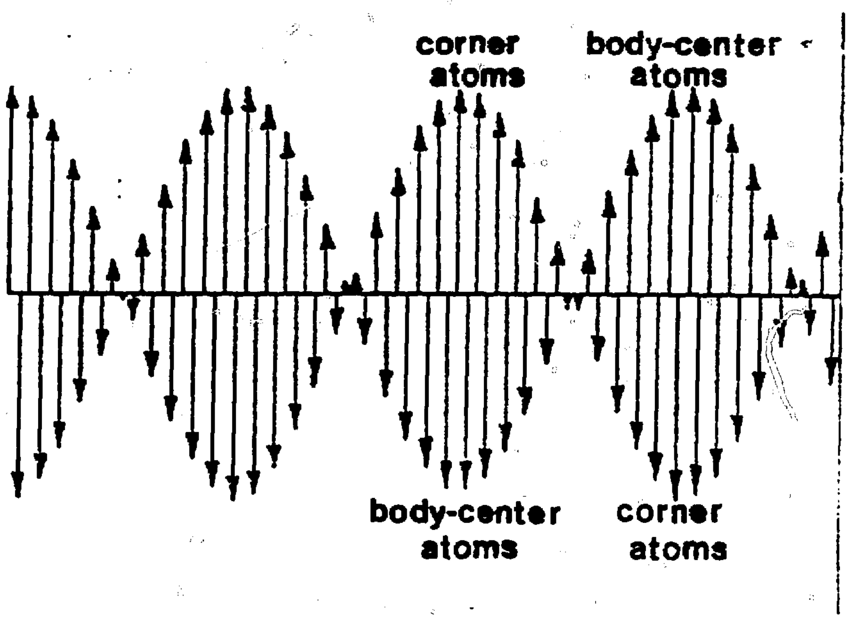

Fig.2. Schematic diagram of the incommensurate magnetization wave in Cr.

domains develop below T_N with the Q vector of the SDW along any one of the three 100 -type directions in the crystal giving rise to three equivalent sets of satellite reflections. It is possible to produce a single-Q state of the crystal by cooling through the Neel temperature in a large applied magnetic field directed along a 100 axis. This procedure results in a crystal consisting of one type of domain with the modulation direction along the applied field. The reciprocal lattice shown in Fig. 1 is for a "field-cooled" crystal with the single-Q direction along 001 . A schematic diagram of the magnetization wave resulting from this SDW state in Cr is shown in Fig. 2.

The magnetic excitations of this static SDW are naturally of great interest and of fundamental importance in magnetism. Theories of the long wavelength spin waves have been developed, based on various simplifying two-band models by Fedders and Martin,[5] Liu[6] and by Sato and Maki.[7] These theories predict the occurence of very high velocity spin waves (comparable to the Fermi velocity). The predictions do not depend strongly on the extent of incommensurability of the SDW. Past measurements on commensurate Cr(Mn) alloys[8] give a spin wave velocity of about 1.5 x 10^7 cm/sec in rough agreement with theoretical predictions. Such high velocities present special problems for typical triple-axis neutron spectroscopy. This paper summarizes the results of our current experimental program aimed at understanding the magnetic excitations occuring in both the incommensurate longitudinal and transverse phases, and in the paramagnetic phase of pure chromium.[9,10]

EXPERIMENTAL RESULTS

The experiments were carried out on triple-axis spectrometers at the Brookhaven High Flux Beam Reactor. Two single crystal samples in the single-Q state were used in the experiments. The most extensive measurements have been made on a highly perfect, vapor-grown single crystal of approximately 0.5 cm^3. We have also taken data on a second larger (2 cm^3), less perfect, crystal grown by the strain and anneal method.

In Fig. 3 we show the results of a constant E (constant energy transfer) scan along 001 across the satellites positions near the (0,0,1) point in reciprocal space. As expected, strong scattering is observed near the satellite Bragg points, from which the high-velocity, probably conical, spin-wave dispersion surfaces originate. Single peaks are observed at .95 $\frac{2\pi}{a}$ and 1.05 $\frac{2\pi}{a}$ because the dimensions of this dispersion surface in momentum space are much smaller than the resolution ellipsoid. In addition to the spin-wave scattering, there is clearly an additional feature centered at the commensurate point (0,0,1). As the temperature is raised the intensity of this diffuse commensurate scattering increases very rapidly as seen in Fig. 3.

In Fig. 4, results of constant-Q scans at the commensurate (0,0,1) point are shown for a series of different temperatures. There is a well defined excitation at about 4 meV at the lower temperatures which shifts to about 3.5 meV at 300K. There is also a dramatic

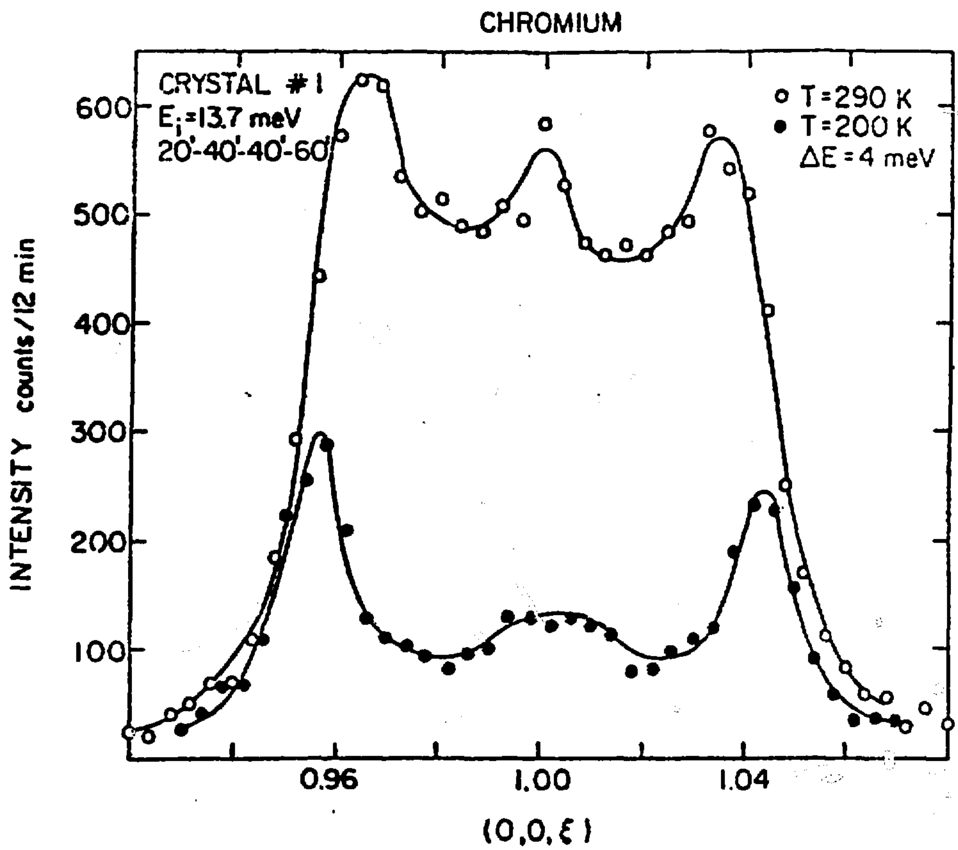

Fig. 3. Constant E scans in the region of the incommensurate magnetic Bragg reflections near (0,0,1). (C.R. Fincher, G. Shirane, S.A. Werner, ref. 10).

increase in the large sloping "background" scattering as the temperature is raised from 130K to 300K. We have carefully repeated this constant-Q scan at 290K and there appears to be an additional peak centered at about 7.5 meV (Fig. 5).

The temperature dependence of this "commensurate-diffuse" scattering is shown for selected energy transfers in Fig. 6. This scattering appears to be diverging at the Néel point. It is, if fact, increasing exponentially as shown by the plot on a log scale shown in Fig. 7. We have made no attempt yet to measure the detailed temperature dependence very close to T_N. This commensurate-diffuse scattering has recently been observed by Mikke and Jankowska[9] in a chromium alloy Cr (.18% Re). They also observe the rapid temperature dependence of this scattering, similar to the data of Fig. 6.

Earlier we had concluded that the fluctuations leading to both the spin wave scattering and this commensurate-diffuse mode were highly anisotropic as a result of a comparison of measurements made near (0,0,1) and (0,1,0).[10] Recently we have found that this

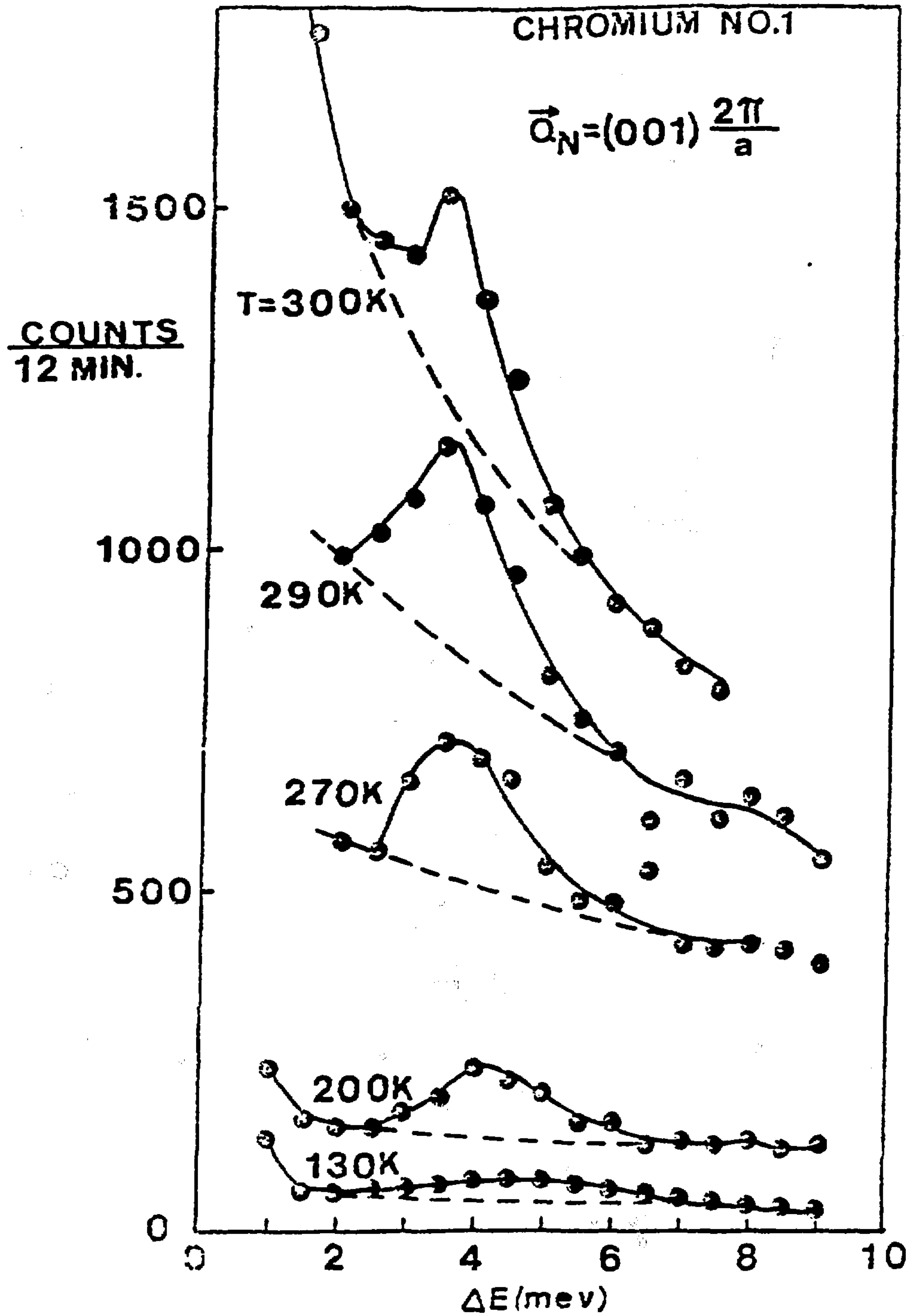

Fig. 4. Constant-Q scans at (0,0,1) at various temperatures showing the energy dependence of the commensurate-diffuse mode. (C.R. Fincher, G. Shirane, S.A. Werner, ref. 10).

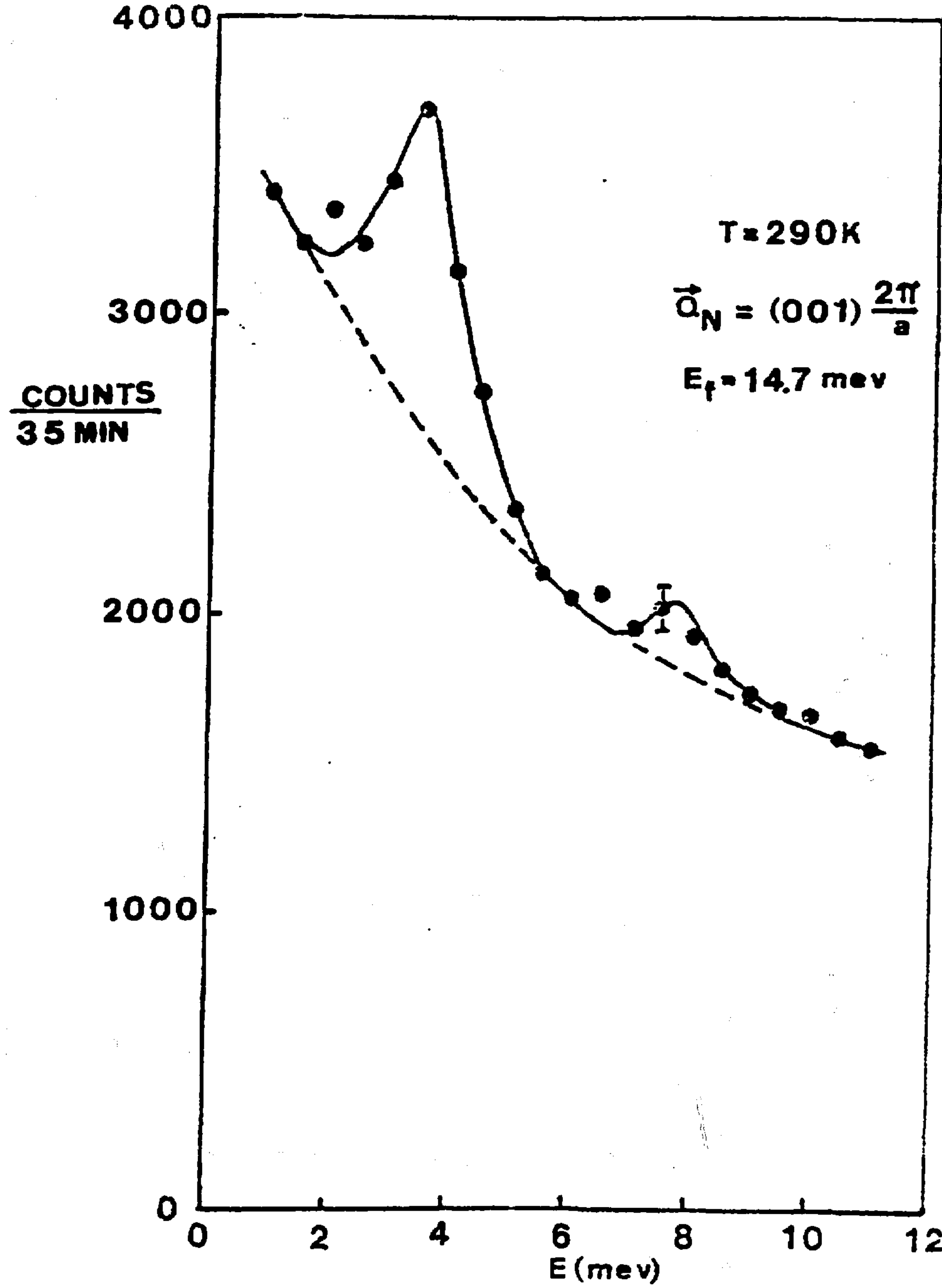

Fig. 5. Constant-Q scan at (0,0,1) at 290K showing an indication of an additional discrete excitation at $E \approx 7.5$ mev. (B.H. Grier, G. Shirane, S.A. Werner, ref. 11).

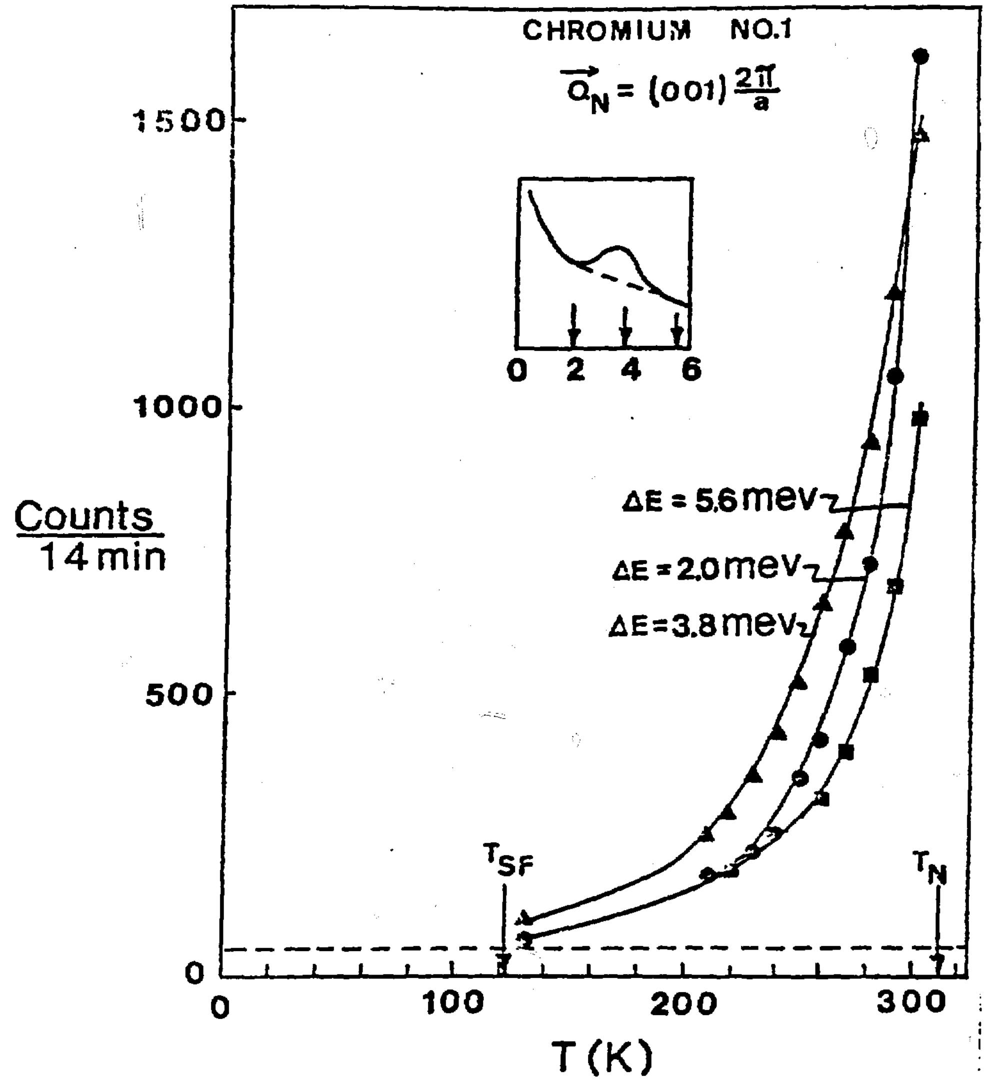

Fig. 6. Temperature dependence of commensurate-diffuse mode at selected energy transfers. (B.H. Grier, G. Shirane, S.A. Werner, ref. 11).

conclusion is incorrect and that the earlier measurements were biased by anisotropic sample capsule transmission effects. Representative scans to resolve this question are shown in the upper portion of Fig. 8 taken at 1[illegible]5K (just above the spin flip temperature). Because the extent of the resolution ellipsoid is large in comparison to the width of the dispersion surface (at these low energy transfers), it is the peak intensities of these scans which should be compared. From a large amount of data of this type we now conclude that the fluctuations

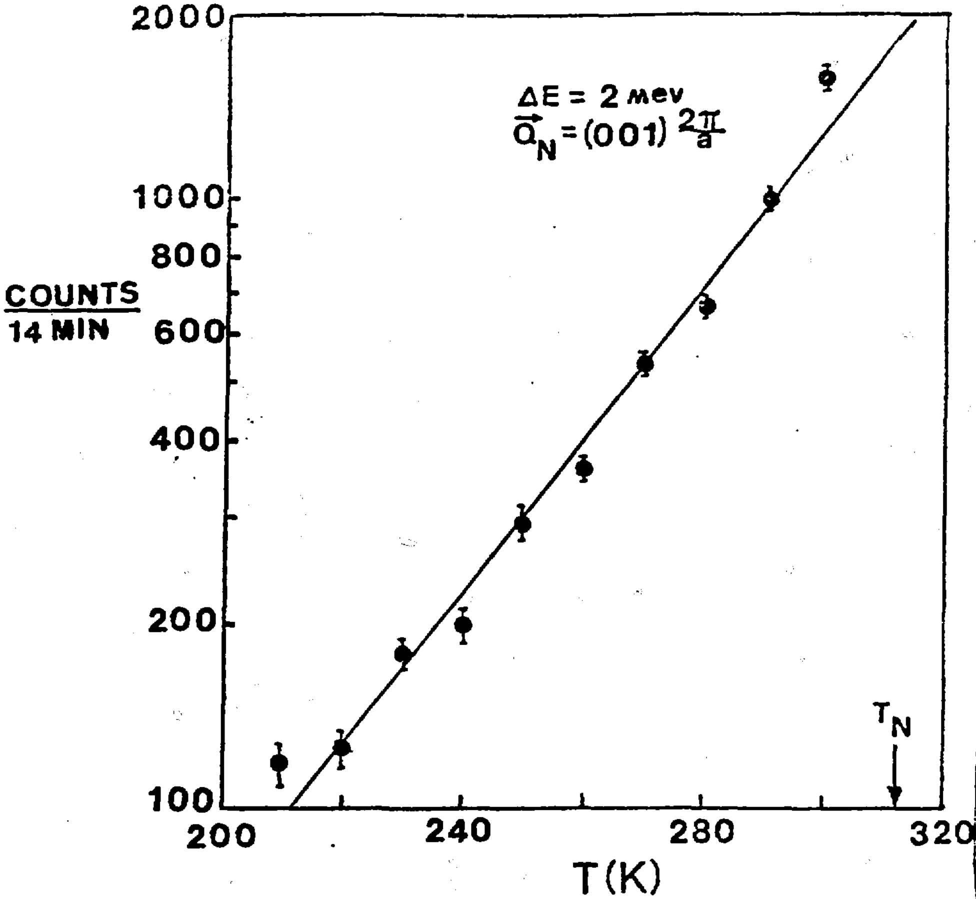

Fig. 7. Temperature dependence of the commensurate excitations at an energy transfer E = 2 mev on a log plot, showing that this intensity increases exponentially with temperature. (B.H. Grier, G. Shirane, S.A. Werner, ref. 11).

leading to the spin wave modes and also to the commensurate-diffuse mode are nearly isotropic in polarization directions. We have not yet resolved the question of whether the spin wave velocity is isotropic.

The data shown in the lower portion of Fig. 8 (taken at 119K, just below the spin flip temperature) is very interesting. The intensity of the scan along [0,0,ξ] is much lower than along [0,1,ξ]. If only transverse fluctuations of the atomic spin were contributing to this scattering one would have expected the reverse intensity ratio. It therefore appears that the longitudianl susceptibility $\chi_{zz}(\vec{q},\omega)$, at least at these values of $\vec{q}$ and ω must be larger than the transverse susceptibility $\chi_{+}^{-}(\vec{q},\omega)$ in the low-temperature longitudinally polarized SDW state. This is a surprising observation.

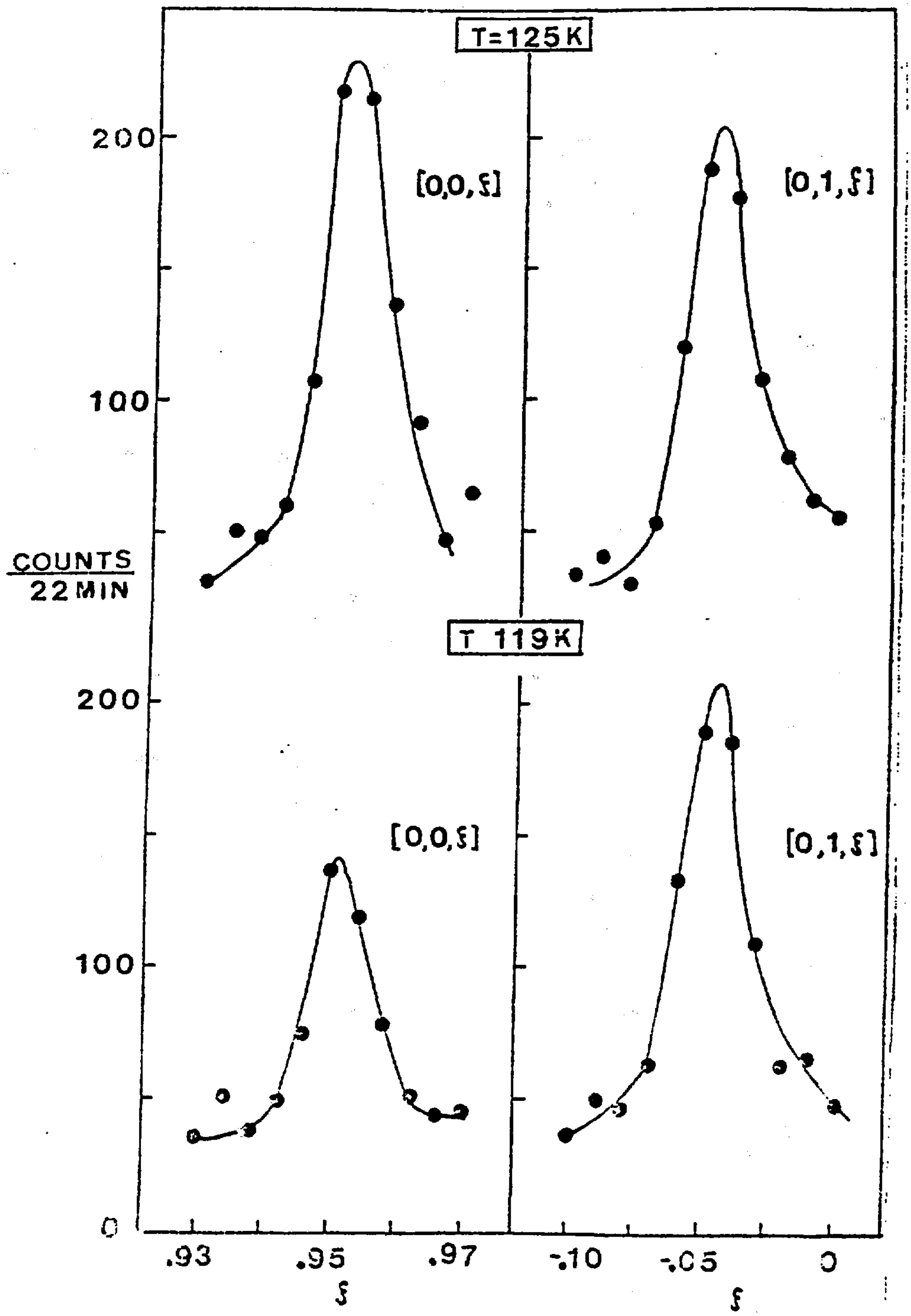

Fig. 8. Constant E (= 4 mev) scans near (0,0,1) and near (0,1,0) just above the spin flip temperature and also just below T_{sf}. (B.H. Grier, G. Shirane, S.A. Werner, ref. 11).

We have carried out experiments at higher energy transfers also, up to ΔE = 30 mev. There are some perplexing features of this data. First of all, the spin wave peaks in the constant-ΔE scans are not centered above the incommensurate Bragg peaks, but are shift inward towards the (0,0,1) point. This means that the dispersion surfaces originating at these Bragg peaks are not simple conical surfaces, described by a single spin wave velocity parameter. Secondly, the intensity in the immediate region around (0,0,1) is considerably stronger than for the lower energy data. The overall trend of a wide assembly of this inelastic data is that as we move toward higher temperatures or toward higher excitation energies, the scattering cross section becomes dominated by the commensurate-diffuse mode.

Recently we have carried out experiments in the presence of a large dc magnetic field (up to 60 KOe). Previous diffraction experiments[12] have shown that the polarization of the spin density wave in the transverse state can easily be rotated in the x-y plane (normal to $\vec{Q}$) by applying a magnetic field $\vec{H}$ normal to $\vec{Q}$. The spin direction tends to allign perpendicular to both $\vec{Q}$ and $\vec{H}$, as is expected in any antiferromagnet where the dc perpendicular susceptibility $\chi_\perp$ is larger than the parallel susceptibility $\chi_\parallel$. Since $\Delta\chi$ $(= \chi_\perp - \chi_\parallel)$ is quite small in Cr, and since a rather modest magnetic field of order 20KOe is sufficient to rotate all of the spins (in the best crystals), the magnetic energies per atom involved in this rotation process are very small. On the basis of these earlier experiments, one would therefore expect low energy transverse (to $\vec{Q}$) fluctuations of the polarization of the SDW. We had thought initially that the commensurate-diffuse scattering might be related to these modes. However, if this were the case, this scattering should be substantially affected by a large magnetic field. We observe no measurable effects of a field on either the commensurate mode or on the spin waves. We find this very surprising, since the field restrains the polarization direction of the static magnetization wave along, say the x-axis; whereas in zero field the spin direction is free to assume any direction in the x-y plane.

We have recently carried out careful experiments with very high energy resolution (down to 20 μV) to look for an energy gap in the spin wave spectrum. We observe well-defined spin waves down to less than 100 μV. This observation also appears surprising, if we think of Cr in terms spin wave theory of conventional, localized moment antiferromagnets in which a gap in the excitation spectrum is related to the exchange interaction and the anisotropy. We know that there is a four-fold anisotropy in the x-y plane from magnetic torque experiments and a very large anisotropy above T_{sf} forcing the polarization of the SDW to be transversely polarized. Since the spin wave velocity is so high, an effective exchange parameter J must also be large.

We show in Fig. 9 constant energy scans, at ΔE = 10 mev, at various temperature in the paramagnetic phase up to temperatures corresponding to about 1.7 T_N. All of this data is similar in shape in momentum space, having a FWHM of $\Delta q \approx .07 \frac{2\pi}{a}$, while simply decreasing monotonically with increasing temperature. The intensity also

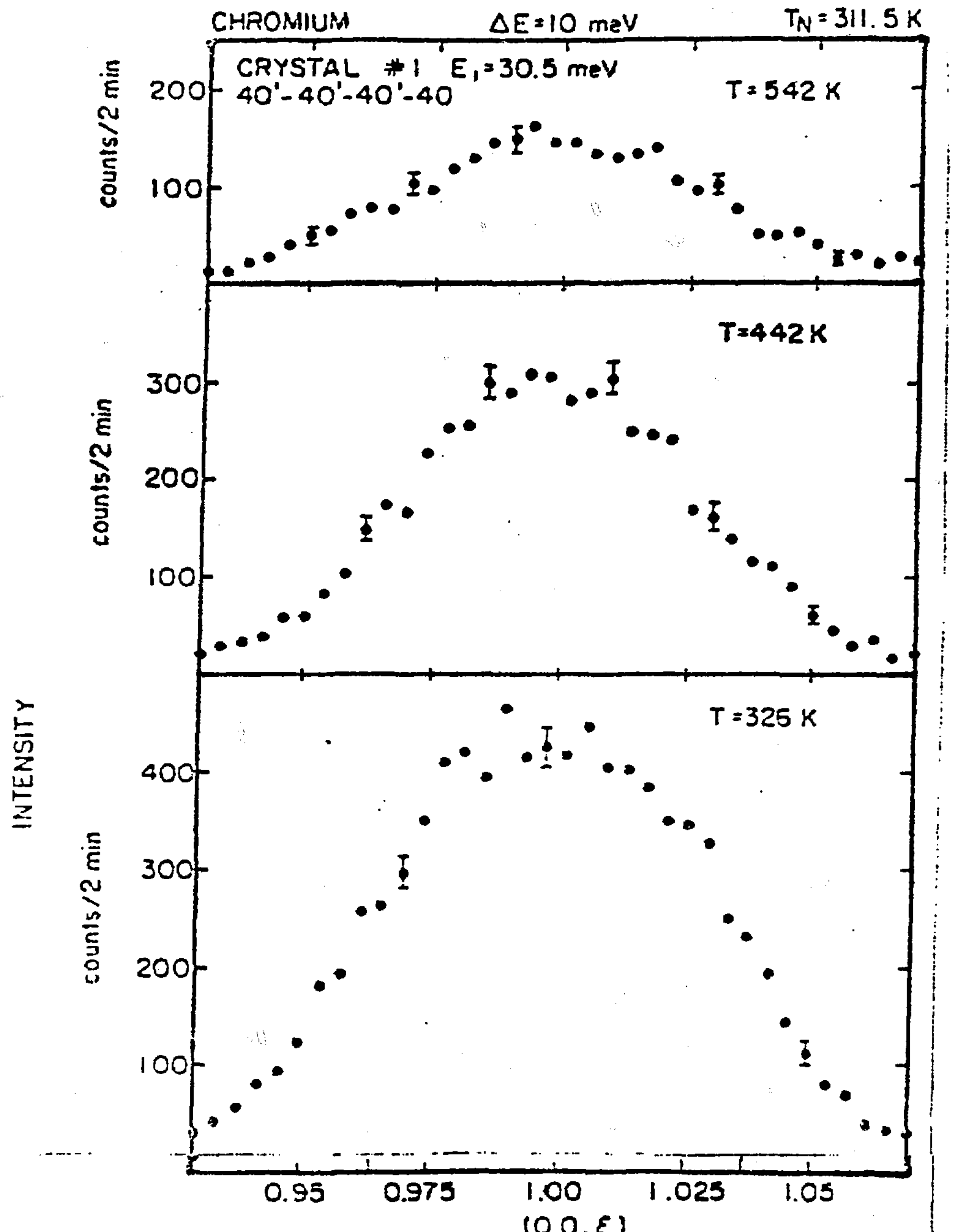

Fig. 9. Inelastic scattering at E = 10 mev at various temperatures in the paramagnetic phase. (C.R. Fincher, G. Shirane, S.A. Werner, ref. 10).

decreases monotonically with increasing energy transfer. From these observations we therefore must conclude that there are well-defined spin-spin correlations extending over 12 to 14 bcc unit cells.

CONCLUSIONS

Although additional experiments are required, especially at higher energy transfers, to understand this evolving picture of the magnetic excitation of chromium, we can already see similarities in these results with other itinerant magnetic systems. The diffuse, reasonably well-localized, scattering in the paramagnetic phase shown in Fig. 9 is qualitatively similar to observations in MnSi, which orders in a helically modulated ferromagnetic structure. This helical structure has a long period of order 150 Å with a $\vec{Q}$-vector in the <111> direction. Thus, the elastic magnetic scattering has a satellite structure, as does Cr. Precursory diffuse scattering qualitatively resembles some of the features of our Cr data. The scattering above T_c is not centered at the satellite positions in reciprocal space but rather around a commensurate Bragg point. As T approaches T_c, the scattering has an approximately spherical shape with a radius comparable to the wavevector distance of the satellites away from the central Bragg point - similar to the data of Fig. 9. The Moriya-Kawabata theory[12] of itinerant magnetism has been successful in understanding some of the results in MnSi, and therefore we might expect it to be helpful in understanding the results on chromium.

It is clear that there are several possible modes of excitations of the static incommensurate SDW in chromium:

1. Ordinarily spin waves which are precessional modes of the 3d atomic moments.
2. Amplitude modes in which the amplitude $\vec{M}_Q$ of the SDW is allowed to fluctuate. These modes contribute to the longitudinal susceptibility $\chi_{zz}(\vec{q},\omega)$ which we apparently are seeing below T_{sf}.
3. Phason modes for which the phase of the SDW as a whole fluctuates, or for which the relative phase of the spin up and spin down electron density fluctuates. This relative phase fluctuation gives rise to dynamic charge density waves.

The relative importance of these three types of excitations in chromium is not known at the present time. Future progress will require a combination of additional experiments and new theoretical insights.

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