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## SPIN WAVES AND THE ORDER-DISORDER TRANSITION IN CHROMIUM

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The inelastic magnetic scattering of neutrons has been studied in Cr and Cr<sub>0.95</sub>-Mn<sub>0.05</sub> both below and above the Néel temperature. The temperature dependence of the spin-wave velocity in the alloy has been measured below  $T_N$ . The scattering above  $T_N$  may also be interpreted in terms of spin-wavelike excitations, which persist far into the paramagnetic phase.

Using a triple-axis spectrometer, we have studied the inelastic magnetic scattering of neutrons in single crystals of Cr and Cr<sub>0.95</sub>-Mn<sub>0.05</sub> both below and above the Néel temperature  $T_N$ . The addition of 5% Mn to Cr causes the spin-density wavelength to become commensurate with the lattice periodicity so that a simple antiferromagnetic structure results.<sup>1</sup> This simplifies the study of the magnetic excitations, and most of the measurements presented in this paper were therefore performed on the alloy.

The inelastic neutron scattering observed in  $\vec{q}$  scans for constant energy transfer  $\hbar\omega$  at room temperature in the alloy is shown in the left of Fig. 1,  $\vec{q}$  being the distance of the scattering vector from (1, 0, 0), which is a point of the magnetic superlattice in reciprocal space. This scattering is interpreted as being of magnetic origin for the following reasons: (i) No peak was observed in  $\vec{q}$  scans through the nonsuperlattice point (1, 1, 0). (ii) A  $\vec{q}$  scan at 10 meV through the (1, 1, 1) superlattice point yielded a width consistent with those of Fig. 1. (iii)  $\vec{q}$  scans through (1, 0, 0) with different spectrometer settings, using various combinations of Be(0, 0, 2), Zn(0, 0, 2), and Ge(1, 1, 1) as monochromator and analyzer planes, all gave widths consistent with those of Fig. 1. (iv) The pronounced temperature dependence of scans at 10 meV through (1, 0, 0), shown in the lower part of Fig. 1, is strong evidence that the scattering is of magnetic origin.

The magnetic excitations in an itinerant anti-

ferromagnet have been discussed by Fedders and Martin<sup>2</sup> and have the form of spin waves with a linear dispersion relation at small wave vectors. We have therefore interpreted our data in terms of undamped spin waves with a scattering cross section of the form  $S(\vec{q}, \omega) \sim \delta(\omega - c|\vec{q}|)$ . Because of the limited resolution of the spectrometer, the peak around (1, 0, 0) is composed of unresolved contributions from spin waves propagating in all directions. In principle it might be possible to resolve these contributions by using larger scattering vectors, e.g., around the (1, 1, 1) reflection, but in practice the decrease in the form factor<sup>3</sup> results in impracticably low intensities. The interpretation of the results therefore depends upon a very careful treatment of the experimental resolution. The intensity  $I(\vec{q}^0, \omega^0)$  at the spectrometer setting  $(\vec{q}^0, \omega^0)$  is the convolution of the experimental resolution function  $R(\vec{q}, \omega)$  and  $S(\vec{q}, \omega)$ , so that

$$I(\vec{q}^0, \omega^0) \propto \int R(\vec{q} - \vec{q}^0, \omega - \omega^0) S(\vec{q}, \omega) d^3q d\omega. \quad (1)$$

We have assumed that the angular distributions of the mosaic blocks in the crystals and the collimator transfer functions may be approximated by Gaussians, and that the component  $q_z$  normal to the scattering plane is uncorrelated with the other variables. If we now approximate the cone dispersion surface by a cylinder within the range of the resolution function, and use the fact that the resolution width in the direction of  $q_z$  is much greater than  $\omega^0/c$ , we find

$$I(q_x^0, 0, 0, \omega^0) \propto \int_{q_x^2 + q_y^2 = 0}^{(\omega^0/c)^2} \left[ \left( \frac{\omega^0}{c} \right)^2 - q_x^2 - q_y^2 \right]^{-1/2} \exp \left[ -\frac{(q_x - q_x^0)^2}{\sigma_x^2} - \frac{q_y^2}{\sigma_y^2} - k(q_x - q_x^0)q_y \right] dq_x dq_y, \quad (2)$$

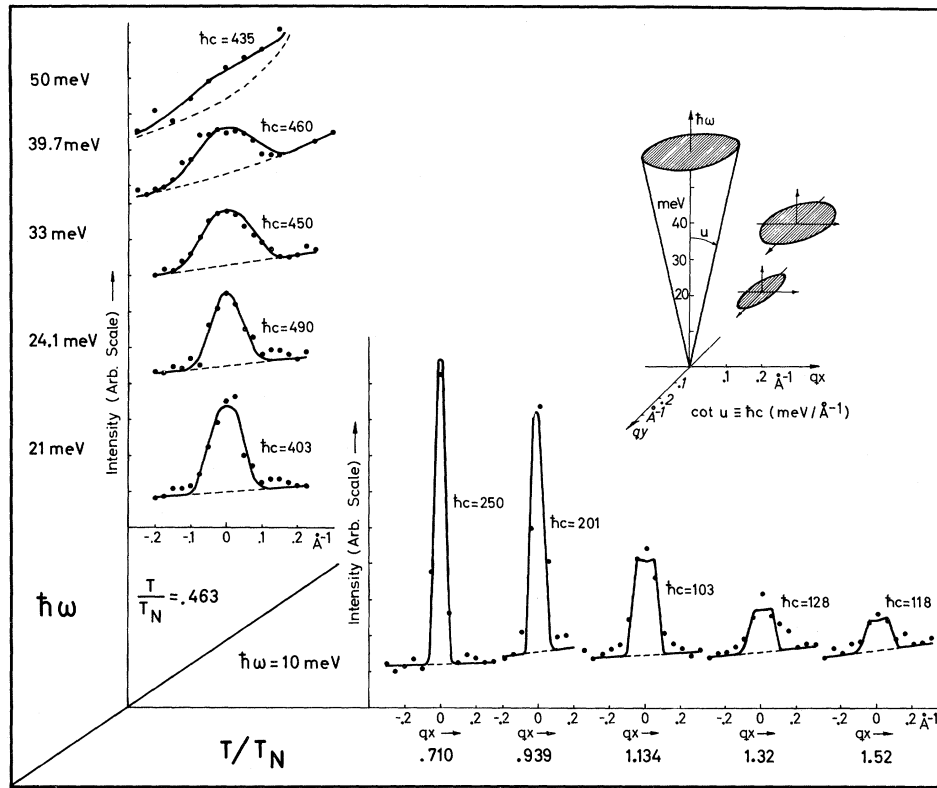


FIG. 1. Intensities from  $\vec{q}$  scans at constant energy transfer for the  $\text{Cr}_{0.95}\text{Mn}_{0.05}$  sample. The widths at five energies at room temperature ( $T/T_N = 0.463$ ) are consistent with the linear spin-wave dispersion relation indicated in the upper right part of the figure. The  $1/e$  contours of the resolution function at 20 and 40 meV are also shown. Filled circles are the measured intensities, the solid lines represent the calculated line profile in a least-squares fit of  $\hbar c$ , and the dashed lines indicate the background. Intensities from identical  $\vec{q}$  scans at 10 meV but with the sample at different temperatures are shown in the lower part of the figure.

where  $\sigma_x$ ,  $\sigma_y$ , and  $k$  all depend on  $\omega^0$ .

The solid lines in Fig. 1 are least-squares fits of Eq. (2) to the data, allowing  $\hbar c$  to vary. The independent determinations of  $\hbar c$  at five energies at room temperature are consistent, and give an average value of about  $450 \text{ meV}/\text{\AA}^{-1}$ . Muhlestein and Sinha<sup>4</sup> have studied magnetic scattering in Cr-Mn alloys by the diffraction method and found initially values of  $220 \text{ meV}/\text{\AA}^{-1}$  for a 1.6% alloy and  $94 \text{ meV}/\text{\AA}^{-1}$  for a 5.7% alloy. A reanalysis of the data by Sinha (private communication) gave a value of  $370 \text{ meV}/\text{\AA}^{-1}$  for the more concentrated alloy, much closer to our result.

The isotropic two-band model of Fedders and Martin, which is based on the Lomer model,<sup>5</sup> gives a value  $c = \bar{v}/\sqrt{3}$  at low temperatures, where  $\bar{v}$  is the geometrical mean of the Fermi velocities in the two bands. For Cr,  $\bar{v}$  may be estimated as about  $4 \times 10^7 \text{ cm/sec}$ ,<sup>6</sup> giving a value of  $1500 \text{ meV}/\text{\AA}^{-1}$  for  $\hbar c$  at  $T = 0$ . This is in reasonable agreement with our result, which is for  $T = 0.5T_N$ . It should be noted that, according to

our analysis, the whole of the magnetic scattering may be ascribed to spin waves, without the necessity of postulating other excitations.<sup>4</sup>

Similar measurements have been carried out above room temperature and, as shown in Fig. 1, the spin-wave velocity appears to decrease almost linearly with temperature as  $T_N$  is approached. There is no dramatic change in the inelastic magnetic scattering on passing through  $T_N$  and, as shown in Fig. 1, it persists to above  $1.5T_N$ . The results in the paramagnetic phase may be consistently interpreted in terms of spin-wave excitations with an almost constant value of about  $100 \text{ meV}/\text{\AA}^{-1}$  for  $\hbar c$ . Further experiments are being performed to elucidate more completely the nature of the magnetic scattering in the paramagnetic phase.

The magnetic transition in the alloy appears to be of second order and the order parameter decreases as  $(T - T_N)^{1/2}$  on approaching the Néel temperature, although the results are somewhat uncertain because of macroscopic concentration

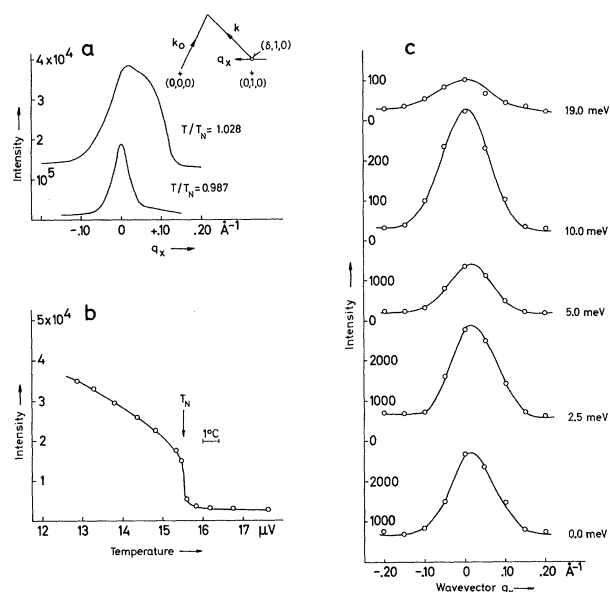


FIG. 2. Intensity data for a pure Cr sample around the  $(\delta, 1, 0)$  satellite. (a) Line profiles measured with a double-axis spectrometer showing the broadening above  $T_N$ . The displacement and distortion of the peak above  $T_N$  is due to overlapping intensities from the other satellites. (b) Intensity versus temperature for the  $(\delta, 1, 0)$  magnetic reflection. (c) Energy analysis of the scattered intensity at  $T/T_N = 1.067$ . Note the change of scale at 10.0 meV.

inhomogeneities in the crystal. In pure Cr, on the other hand, the transition is of first order, as first pointed out by Arrott, Werner, and Kendrick.<sup>7</sup> The crystal used for our investigation was the same as that used by Møller *et al.*<sup>8</sup> in their original study of magnetic scattering in the paramagnetic phase of Cr and, as illustrated in Fig. 2(b), also shows a first order transition. The measurements of Møller *et al.*<sup>8</sup> were performed on a double-axis spectrometer, as were those of Hamaguchi, Tsunoda, and Kunitomi.<sup>9</sup> We have also carried out such a measurement, as shown in Fig. 2(a), and in addition we have performed an energy analysis of the scattered

beam. The results for  $T/T_N = 1.067$  are shown in Fig. 2(c). A quantitative interpretation of these results is precluded by the complexity of the magnetic structure, which now produces six satellite reflections about  $(1, 0, 0)$ , but at room temperature, corresponding to  $T/T_N = 0.94$ , the observed widths around the  $(\delta, 1, 0)$  satellite were  $0.12 \text{ \AA}^{-1}$  at  $\hbar\omega = 10 \text{ meV}$  and  $0.15 \text{ \AA}^{-1}$  at 19 meV. These figures indicate that  $\hbar c$  has the same order of magnitude as in the alloy. The strong inelasticity of the magnetic scattering in the paramagnetic phase makes it clear that it is unjustified to analyze the results in terms of a quasistatic model, as was done by Møller *et al.*,<sup>8</sup> but their conclusion that strong dynamic fluctuations in magnetization occur far into the paramagnetic phase remains valid.

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