Spin density wave behaviour in the \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\) and \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) alloy series

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Abstract. Neutron diffraction studies of the spin-density-wave (SDW) behaviour in antiferromagnetic \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\) with \(y = 0, 3.7\) and 4.2, as well as \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) with \(x = 0\) and 4.6 alloy systems are reported. The results confirm an incommensurate (I) SDW to paramagnetic (P) phase transition at the quantum critical point of the \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\) series. In addition two further quantum critical magnetic phase transitions are proposed for the \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) series, one from ISDW to P and the other from P to commensurate (C) SDW.

1. Introduction
The combination of properties of Cr and its dilute alloys associated with the Fermi surface, magnetic structure and spin-density-wave (SDW) state provides for an ideal case-study of itinerant electron antiferromagnetism (AFM) in metals [1]. Alloying Cr with Al, a group 3 non-transition metal element, decreases the Néel temperature \((T_N)\) reaching a minimum near 2 at.% Al, where after \(T_N\) increases on further Al addition [1, 2, 3]. This minimum occurs at a critical concentration where the incommensurate (I) SDW, commensurate (C) SDW and paramagnetic (P) phases co-exist [2, 3] on the magnetic phase diagram (MPD). Recent research suggests that this triple point might indeed be quantum critical (QC) in nature [3].

Addition of 5 at.% Mo to the \(\text{Cr}_{100-x}\text{Al}_x\) alloy system completely suppresses AFM in the concentration range \(2 \leq x \leq 5\), after which it reappears on further Al addition [1, 4]. This results in two possible quantum critical points (QCP) on the MPD of the \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) system, one expected to be an ISDW-P and the other a P-CSDW phase transition [5]. On the other hand, the \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\) series indicate only one QCP, expected to be an ISDW-P transition [6, 7].

In the present study, neutron diffraction measurements have been performed to confirm the nature of the magnetic phases around the QCPs predicted from previous studies of \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) [1, 4, 5] and \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\) [6, 7] alloys.

2. Experimental
Two sets of polycrystalline ternary \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\) with \(y = 0, 3.7\) and 4.2, and \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) with \(x = 0\) and 4.6 alloys were prepared as was previously described [5, 6, 7]. Analyses confirmed that the samples were homogeneous, with actual concentrations of \((1.6 \pm 0.1)\) at.% Al in the \((\text{Cr}_{98.4}\text{Al}_{1.6})_{100-y}\text{Mo}_y\), and \((5.0 \pm 0.2)\) at.% Mo in the \((\text{Cr}_{100-x}\text{Al}_x)_{95}\text{Mo}_5\) series, respectively. Temperature dependence
of the electrical resistivity (ρ) and magnetic susceptibility (χ) measurements gave TN values corresponding with that previously obtained [5, 6, 7]. Investigations were extended to neutron diffraction measurements at the Australian Nuclear Science and Technology Organisation (ANSTO) using the Wombat powder diffractometer [8].

3. Results and discussion
Examples of neutron diffraction patterns for the (Cr_{98.4}Al_{1.6})_{100-x}Mo_{x} and (Cr_{100-x}Al_{x})_{95}Mo_{5} alloys taken at the (100) reciprocal lattice positions (rlp) are shown in figure 1. The presence of the diffraction peaks are purely of magnetic origin [1]. Diffraction patterns were obtained at various set temperatures on cooling from the P phase of each alloy to 4 K. A monochromatic wavelength of 2.41 Å was used for the sample with y = 0, with 1.54 Å for the samples with x = 0 and 4.6. Magnetic satellites at the (1±δ,0,0) rlp for the alloys with y = 0 and x = 0 indicate that these alloys order in the ISDW phase [1, 2]. In this phase the magnetic moments are perpendicular to the wave vector Q directed along the (100) cubic axis, given by [1, 2]: \( Q = \frac{2\pi}{a} (1 \pm \delta, 0, 0) \), where \( a \) is the lattice parameter and \( \delta \) is a parameter that measures the deviation from a CSDW state. Figure 1(c) shows a weak single diffraction peak at the (100) rlp for the x = 4.6 alloy. The absence of magnetic satellites indicates that this alloy orders in the CSDW phase, i.e. \( \delta = 0 \). Alloys with y = 3.7 and 4.2 did not show magnetic reflections, despite the presence of a low temperature minimum in the ρ(T) curves. These concentrations are close to the expected QCP [6], which leads to weak intensities that were possibly indistinguishable above the background.

Figure 2 shows representative graphs of the temperature dependences of the integrated neutron intensities. Results of the solid rod and powdered samples for the alloy with y = 0 are shown in figure 2(a), with figure 2(b) showing that of the solid rod sample with x = 0. Typically, the integrated neutron intensities decrease with temperature, levelling-off in the P region. TN was subsequently taken at the temperature where a quadratic fit to the data points in the AFM phase intersects with a linear fit in the P phase. For the x = 0 sample the TN-value corresponds well with that obtained from ρ(T) and χ(T) measurements, despite the limited number of points in the P phase. TN-values obtained for the powdered and solid rod samples for the y = 0 alloy are similar. Surprisingly it indicates that crushing the sample didn’t affect TN significantly, contrary to results previously reported for pure Cr [9, 10].

Investigations of the alloy with x = 4.6 at several set temperatures revealed the presence of a central peak of magnetic origin at 4 K and 25 K, that was absent for \( T \geq 50 \) K. With the limited number of data points in the AFM phase, the temperature dependence of the integrated intensity curve could not be plotted. It should be noted that the neutron intensities in the CSDW are weaker than in the ISDW phase, as was also observed in other experimental results on these alloys [3, 5].

![Figure 1](image_url)

**Figure 1.** Neutron diffraction patterns for the antiferromagnetic (Cr_{98.4}Al_{1.6})_{100-x}Mo_{x} and (Cr_{100-x}Al_{x})_{95}Mo_{5} alloys with (a) y = 0 (rod), (b) x = 0 (powder) and (c) x = 4.6 (powder). The temperatures at which the patterns were taken are indicated in each panel.
4. Conclusions

$T_N$-values obtained from $\rho(T)$, $\chi(T)$ and neutron diffraction measurements for each of the samples with $y = 0$ and $x = 0$ are comparable. No additional intensity anomalies that can be attributed to magnetic changes were observed in the neutron diffraction results of the samples with $y = 3.7$ and $4.2$ despite low temperature minima observed in $\rho(T)$ curves. $T_N$ for the alloy with $x = 4.6$ could not be determined from the neutron diffraction measurements since measurements were only taken at two temperature set points in the AFM phase. Previous [5] electrical transport and specific heat measurement indicated that the AFM behaviour on the MPD of the (Cr$_{100-x}$Al$_x$)$_{95}$Mo$_5$ alloy system is fully suppressed down to 2 K in the concentration range $2 < x < 4.4$, after which it then reappears at larger $x$. The present neutron diffraction data indicates that an alloy with $x = 4.6$ orders in the CSDW phase for $T \leq 25$ K. The concentration of this alloy lies close to the second critical concentration $x = 4.4$ [5] where AFM reappears on the MPD of this system. This is a first clear indication that AFM in this alloy system reappears in the CSDW phase. The results furthermore confirm an ISDW-P phase transition, previously inferred from $\rho(T)$ and $\chi(T)$, at the QCP of the (Cr$_{98.4}$Al$_{1.6}$)$_{100}$Mo$_5$ series [6]. Integrated intensities as a function of temperature were measured for the $y = 0$ alloy, both in powder and solid state form, to obtain $T_N$ for this alloy. Contrary to expectation, crushing of the $y = 0$ sample was not found to influence $T_N$ significantly.

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