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Local magnetic moments in dilute Cr–Nb alloys: the effects of applied magnetic field and Nb concentration

P E N de Souza[1], **L M de Oliveira**[1], **W A Ortiz**[1], **P C de Camargo**[2] **and A J A de Oliveira**[1,3]

[1] Departamento de Física, Universidade Federal de São Carlos, CP 676, 13565-905, São Carlos SP, Brazil
[2] Departamento de Física, Universidade Federal do Paraná, CP 19091, Curitiba, PR, 81531-990, Brazil

E-mail: adilson@df.ufscar.br (A J A de Oliveira)

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Abstract
In this work we present magnetic susceptibility results for Cr–x at.% Nb alloys ($x = 0.2$, 0.6, 0.7, 1.4, and 2.0), showing that a local short-range order spin-density wave (L-SDW) appears at a characteristic temperature (T_{loc}) above the Néel temperature. The evidence for L-SDW is based on a Curie–Weiss-like behaviour, which is suppressed when large magnetic fields are applied or for alloys with Nb concentration above $x = 2.0$ at.%.

1. Introduction

Chromium is the best example of a spin-density wave (SDW) itinerant antiferromagnet. The ordering phase occurs at the Néel temperature (T_N) of 311 K. The wavevector Q associated to the SDW is incommensurate with the reciprocal lattice, $Q = 2\pi/a(1 + \delta)$, where δ is the incommensurability parameter. The persistence of this magnetic state in Cr alloys over a wide range of compositions, in particular with transition metals, opens interesting possibilities for magnetic studies in these alloys. The introduction of small quantities of other transition metals in chromium such as V, Ti and Nb decreases the electron/atom ratio, causing an increase in the incommensurability parameter δ, and decreasing the Néel temperature (T_N). On the other hand, the introduction of elements such as Mn, Re, and Fe increases the electron/atom ratio, increasing T_N and decreasing δ [1]. For 0.1 at.% Mn in Cr, δ vanishes.

The introduction of ferromagnetic elements in Cr, may lead to a Curie–Weiss-like (CW-L) behaviour both in the antiferromagnetic and in the paramagnetic phases, as in the case of Cr–Fe [2, 3] and Cr–Mn [4] alloys. This behaviour is associated to intrinsic local magnetic moments presented by these elements. However, there is a possibility of formation of local magnetic moments in Cr alloys with non-magnetic elements as is the case of dilute V in Cr,

[3] Author to whom any correspondence should be addressed.

where CW-L behaviour is observed within a certain temperature range above T_N [5, 6]. This behaviour is suppressed in either of the following cases: for V concentrations above 0.4 at.% V, due to high magnetic fields (10 kOe) [7, 8], or for temperatures higher than a value called local magnetic ordering suppression temperature T_{LOC} [9]. This characteristic temperature T_{LOC} also exhibits a strong dependence on magnetic field and V concentration.

An approach to explain this special magnetic behaviour in Cr with non-magnetic impurities was proposed by Tugushev [10], whose model assumes the establishment of local spin-density waves around lattice or chemical defects. Tugushev's model predicts that this local SDW may be stable even for temperatures higher than T_N. The introduction of impurities or defects in these alloys may build a local rearrangement of the spin-density wave lowering the electrostatic energy. This condition may lead to a self-consistent redistribution of the charge and spin densities around their vicinities, lowering the electrostatic energy and thus leading to the formation of a local spin-density wave (L-SDW) with a CW-L response. Tugushev's model has successfully described the magnetic behaviour in several situations such as a CW-L response in Cr–V alloys [5–9], magnetic frustration above T_N in Cr–Fe–V alloys [11] and Cr–Co–V [12]. In all these cases, V impurities produce thermomagnetic irreversibilities above T_N associated to local magnetic moments with short-range order and magnetic frustration.

Although the CW-L behaviour has been extensively studied in recent years [5–12], there still are many open questions, such as those of how impurities interfere in magnetic order. It is thus desirable to study, in a systematic manner, the effects on the magnetic response due to different non-magnetic impurities diluted in Cr. This paper, which is part of such a comprehensive study, reports on the observation of L-SDW in Cr–Nb alloys. The CW-L behaviour and its dependence on magnetic field and on Nb concentration for Cr–x at.% Nb alloys ($x = 0.2$, 0.6, 0.7, 1.4, 2.0) are shown. The magnetic behaviour is very similar to that of Cr–V alloys, supporting the idea that local electronic disturbance is the main origin of such CW-L behaviour. However, a few features clearly distinguish both systems.

2. Experimental details

Polycrystalline samples of Cr–Nb were prepared in an arc-melting furnace, using high-purity Cr (99.9999%) and Nb (99.99%). Each sample was melted several times with a mixing procedure to ensure high-quality homogenous alloys with only a few grains. The Néel temperature was determined by AC magnetic susceptibility (χ_{AC}) measurements carried out with a PPMS 6000 (Physical Properties Measurement System, Quantum Design Corporation), with AC magnetic field of 10 Oe and 100 Hz frequency. Values of T_N determined by χ_{AC} are equivalent to those obtained by AC electrical resistivity [6, 13]. All samples exhibit a sharp Néel transition, within a few degrees, which is an indication of good homogeneity, since T_N is very sensitive to concentration.

DC magnetic susceptibility measurements, $\chi_{DC}(T) = M/H$, were performed in a Quantum Design SQUID magnetometer, model MPMS-5S. Measurement followed this protocol: (i) cooling at zero field; (ii) field applied and measurement on warming (zero field cooling, ZFC, $\chi_{ZFC}(T)$); (iii) measurement on cooling (field cooling, FC, $\chi_{FC}(T)$). For ZFC susceptibility measurement the sample is cooled down to 1.8 K at zero magnetic field. When the temperature has stabilized, the magnetic field is applied and the measurement performed, while increasing the temperature at a constant rate (2 K min^{-1}) up to 400 K. After this measurement, the FC process is started, in which the sample is cooled down at 2 K min^{-1} with the same field applied as in the previous procedure. To eliminate any remaining magnetic history, the sample was first heated up to 400 K. Each value of magnetic moment results from an average of two scans taken over a 3 cm length excursion of the sample through the SQUID sensor.

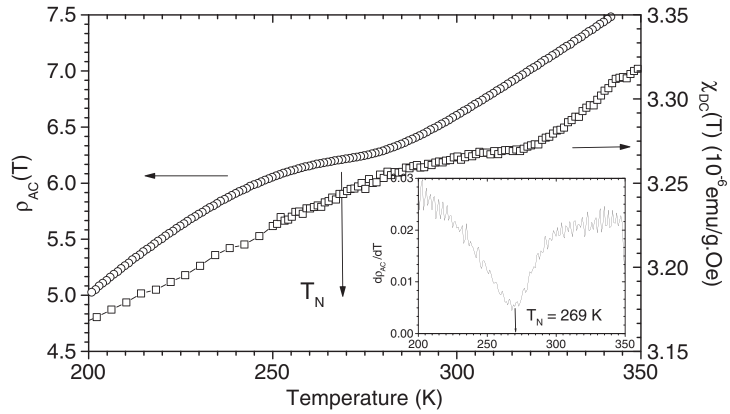

Figure 1. Electrical resistivity, determined by the reciprocal of χ'' and DC magnetic susceptibility (measured at 0.05 Oe) for Cr–0.6 at.% Nb. The inset in the figure shows the slope of the AC electrical resistivity ratio.

3. Results and discussion

The exact determination of T_N in Cr–Nb alloys is very important for the discussion of $\chi_{DC}(T)$ measurements. Figure 1 shows curves of the resistivity ratio, $\rho_{AC}(T)$ (normalized to 1 for $T = 5$ K) and of $\chi_{DC}(T)$ for a Cr–0.6 at.% Nb sample. The reciprocal value of the quadrature signal (χ'') is proportional to $\rho_{AC}(T)$, as demonstrated by de Faria *et al* [13] in the limit of low frequencies. The resistivity curve shown here is identical to those obtained by the four-terminal method [1]. The Néel temperature has been determined, as usual [1, 14], as the inflection point of the resistivity curve, that occurs just below its minimum, as shown in the inset of figure 1. For this sample $T_N = 269$ K. Therefore, the Néel temperature when observed in $\chi_{DC}(T)$ corresponds to a temperature just below the change of slope. In this measurement a magnetic field of 50 Oe was used.

Figure 2 shows the temperature dependence of $\chi_{DC}(T)$ for Cr–0.7 at.% Nb, at fields (a) $H = 0.05$ kOe, (b) 0.5 kOe, (c) 5 kOe, (d) 10 kOe. Panels (a) and (b) show that $\chi_{ZFC}(T)$ increases when the temperature is raised up to 290 K, in the paramagnetic phase ($T_N = 262$ K). At $T = 313$ K $\chi_{ZFC}(T)$ presents a minimum. Above this temperature $\chi_{ZFC}(T)$ increases again. In the FC procedure, we observed that $\chi_{FC}(T)$ is different from $\chi_{ZFC}(T)$, i.e., irreversibility is present at the temperature interval of interest. This behaviour decreases with increasing magnetic fields, being completely suppressed for 10 kOe (panel (d)), for which the system has the typical magnetic behaviour of Cr alloys without any contribution attributable to local magnetic moments. Therefore, field cooling measurements reveal that the L-SDWs are pinned around Nb atoms. As in other reports about Cr alloys which exhibit this behaviour [11, 12], we verified that a CW-L behaviour is observed when the FC and ZFC are different to each other. The L-SDW state appears in ZFC measurements only above the Néel temperature, while it is observed in all range of temperatures in the FC procedure. In fact, when the long-range order (SDW state) disappears, there is a favourable condition for the establishment of local order. When the sample is cooled through its Néel temperature in the presence of a low applied magnetic field, the L-SDW is pinned around defects. In other words, the FC procedure reveals the presence of an L-SDW.

This anomaly observed in the $\chi_{FC}(T)$ curve for the FC state corresponds to the onset of a local-SDW state, as predicted by Tugushev [10]. In this model the introduction of impurities

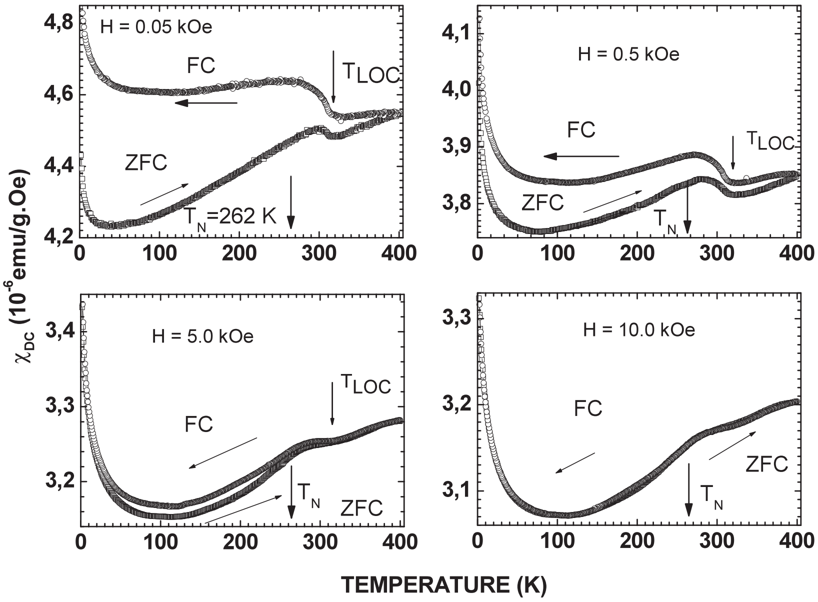

Figure 2. DC magnetic susceptibility for Cr–0.7 at.% Nb. (a) $H = 0.05$ kOe; (b) $H = 0.5$ kOe; (c) $H = 5$ kOe; and (d) $H = 10$ kOe.

or defects produces short-range order regions that develop for $T > T_N$. This behaviour is manifested up to a characteristic temperature, T_{LOC}, at which the order is suppressed. In the present case, $T_{LOC} = 320$ K. Above this temperature, the $\chi_{DC}(T)$ curves show a characteristic behaviour of electronic paramagnetism (Pauli paramagnetism) as observed in pure Cr and other alloys without local magnetic moments. As observed in figure 1, T_{LOC} is not dependent on the applied magnetic field, in contrast to what was previously observed in Cr–V alloys [9], where T_{LOC} exhibits a linear dependence on the applied magnetic field. In similar manner, the suppression of irreversibility due to magnetic field was already found for Cr–Fe–V and Cr–Co–V alloys [14].

Figure 3 presents the effects of Nb concentration on the establishment of an L-SDW. $\chi_{DC}(T)$ measurements were performed at a fixed field of 0.1 kOe for the other alloys studied here, Cr–x at.% Nb ($x = 0.2$, 0.6, 1.4 and 2.0). We found that for $x = 0.2$, 0.6, 1.4 the magnetic behaviour is similar to that of figure 2 where $x = 0.7$. However, for $x = 2.0$, the CW-L behaviour is totally suppressed, even at low magnetic fields. In the case of Cr–V alloys this behaviour is suppressed for lower concentrations (0.67 at.% V) [8]. The defect associated with Nb produces an L-SDW stable up to 290 K, which is well above the Néel transition; however, the local magnetic moment disappears within a few degrees, in contrast to Cr–V alloys, where the L-SDW persists 100 K above T_N [9]. On the other hand, magnetic fields of 1 kOe are enough to suppress the CW-like behaviour for Cr–Nb, while for Cr–V, 10 kOe are needed to obtain similar results.

The differences between the magnetic behaviour in the paramagnetic phase of Cr–V and Cr–Nb may be understood considering that Nb has wider d- and s-band widths and larger atomic radius. According to Tugushev's model, the appearance of L-SDW around defects creates a region of nonzero magnetization, M, where short-range magnetic order takes place. To first order, if one assumes F_1 as the defect potential, $N(0)$ as the density of states at the Fermi level, and Δ as an order parameter proportional to the energy gap, the contribution of

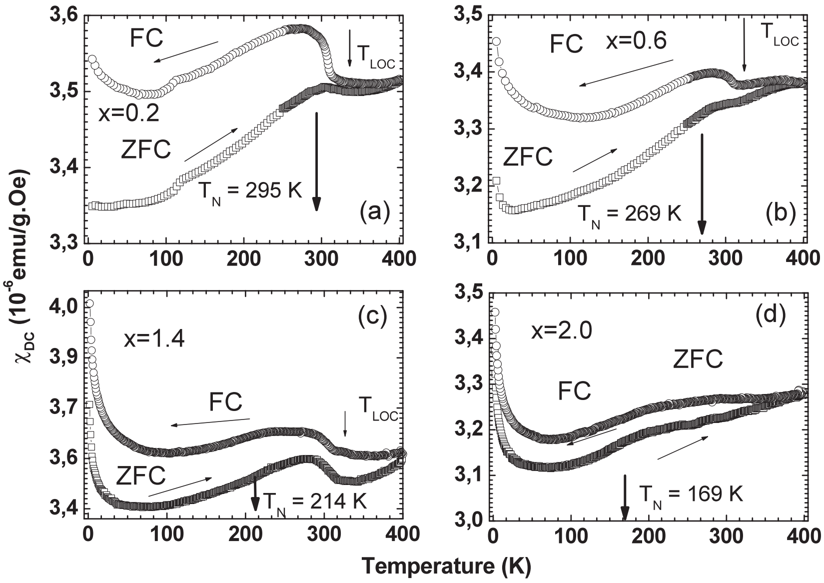

Figure 3. DC magnetic susceptibility for Cr–x at.% Nb, at $H = 0.1$ kOe. (a) $x = 0.2$; (b) $x = 0.6$; (c) $x = 1.4$; and (d) $x = 2.0$.

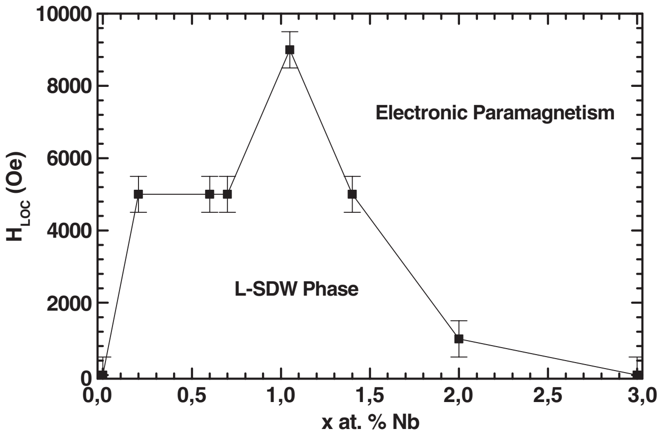

Figure 4. Magnetic phase diagram of the characteristic magnetic field H_{LOC} versus Nb concentration. The error bars represent the uncertainty in H_{LOC} values. The straight line is only a guide to the eye.

magnetization associated to the L-SDW is given by $M = 2F_1\mu_B N(0)\Delta(0)$ [10]. Since all these parameters depend on the type of dopant, the fact that Nb contributes with 4d electrons seems to be an important difference when compared to V.

As the appearance of local spin-density waves depends on Nb concentration and on the applied magnetic field, one can sketch a magnetic phase diagram, showing the boundary line $H_{\mathrm{LOC}}(x)$ separating the L-SDW phase from the electronic paramagnetic phase. Figure 4 represents such a schematic phase diagram. The critical magnetic field was determined as the lowest value for which Pauli paramagnetism behaviour is observed. In other words, above this limiting field the magnetic susceptibility increases with temperature for $T > T_N$. This magnetic phase diagram is very similar to those of Cr–V alloys, although the range of applied magnetic fields is ten times smaller [7].

4. Conclusion

One may then conclude that Cr–Nb alloys, similarly to Cr–V alloys, also present local spin-density wave order above T_N, which can be suppressed by a sufficient increase of the Nb concentration, large enough magnetic fields, and sufficiently high temperatures. Even though the magnetic characteristics are quite different in these systems, both present a CW-L behaviour and the L-SDW state is suppressed in high magnetic fields. In addition, our results support the prediction of Tugushev's model that L-SDWs are formed around impurities in Cr alloys, stimulating the search for similar magnetic behaviour in other Cr alloys, particularly for systems with small impurity concentrations submitted to low applied magnetic fields.

Acknowledgments

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