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LETTER TO THE EDITOR

A new magnetic phase in the SDW alloy Cr + 3.2% Co: effect of doping with V

V Yu Galkin†§, W A Ortiz† and Eric Fawcett‡

† Departamento de Fisica, Universidade Federal de São Carlos, Brazil
‡ Physics Department, University of Toronto, Toronto, Canada

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Abstract. When Cr + 3.2% Co is field cooled (FC) from 400 K through the Néel temperature $T_N \sim 330$ K in the 100 Oe measuring field of a SQUID magnetometer, irreversibility of the magnetic susceptibility $\chi(T)$ occurs relative to the zero-field-cooled (ZFC) state, beginning at the irreversibility limiting temperature $T_i = 385$ K. $T_i(x)$ decreases in ternary (Cr + 3.2% Co)$_{1-x}$V$_x$ alloys with up to $x = 1\%$ V, but more slowly than $T_N(x)$, and the relative magnitude of the hysteresis increases. It is believed that a phase transition occurs at T_i, perhaps to a local spin-density-wave state.

The occurrence of frustration manifested by irreversibility of the magnetic susceptibility $\chi(T)$ of the field-cooled (FC) state relative to the zero-field-cooled (ZFC) state in spin-density-wave (SDW) alloys of Cr containing Mn, both binary Cr$_{1-x}$Mn$_x$ [1, 2] and ternary CrSiMn [1] and CrFeMn [3], led us to explore SDW Cr alloys with other 3d metals that form a magnetic moment when dissolved in Cr. In fact, Fe is the only solute atom that gives rise to a temperature dependence of the magnetic susceptibility in accordance with the Curie–Weiss (C–W) law, in both the paramagnetic and the SDW phase [4, 5]. We have found, however, that neither binary CrFe nor ternary CrVFe [6, 7] alloys show frustration at low concentrations of only a few per cent Fe.

Cr$_{1-x}$Co$_x$ alloys, on the other hand, at low concentrations exhibit a C–W law in the paramagnetic phase corresponding to an effective moment of about $2\mu_B$ [4, 8], but not in the SDW phase [5]. We report here anomalous behaviour that we have observed in the temperature dependence of the susceptibility $\chi(T)$ for a binary Cr + 3.2% Co alloy, and for ternary alloys of compositions (all in at.%), (Cr + 3.2% Co)$_{1-x}$V$_x$: $x = 0.1$, 0.3, 0.4 and 1.0% V. The experimental procedure and methods of sample preparation and characterization are the same as those followed in [1, 2].

The magnetization M was measured with a SQUID magnetometer MPMS-5S from Quantum Design in San Diego. The temperature dependence of magnetic susceptibility, defined as $\chi(T) = M(T)/H$, was measured in the range $5 \leqslant T \leqslant 400$ K, with increasing temperature T after cooling in zero magnetic field H (the ZFC state), and then with both decreasing and increasing T after cooling in the measuring field of 100 Oe (the FC state). Although relaxation effects were not studied systematically, it is worth mentioning that the temperature sweeps normally took about 4 h.

The binary Cr + 3.2% Co alloy shows divergence starting at $T_i = 385$ K between the ZFC and both the FC curves of $\chi(T)$, as illustrated in figure 1. We have found that binary

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§ On leave from: I P Bardin Central Research Institute for Ferrous Metallurgy, 2 Baumanskaya 9/23, Moscow, 107055 Russia.

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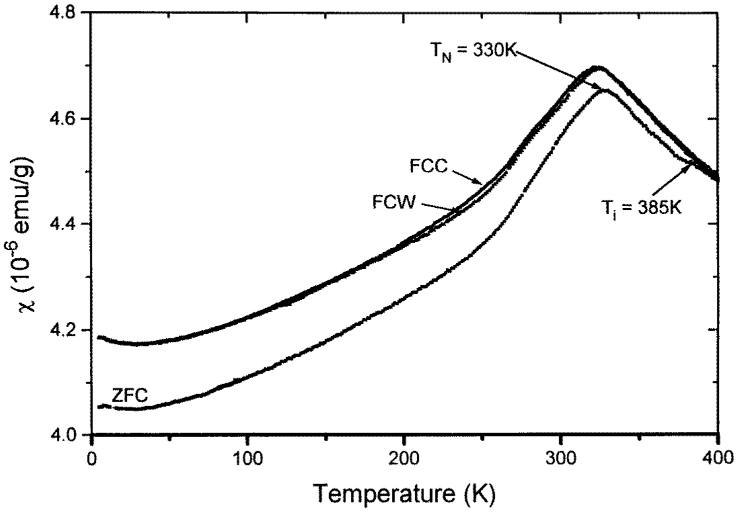

Figure 1. Temperature dependence of the magnetic susceptibility of the binary alloy Cr + 3.2% Co measured in a field of 100 Oe after zero-field cooling (ZFC), and in the same field while cooling from a temperature above 400 K (FCC) and then warming (FCW). The Néel temperature, as determined by the peak in the ZFC curve, and the irreversibility limiting temperature T_i, are indicated.

SDW CrFe alloys show no sign of frustration [6], but adding V gives effects similar to those reported here [7]. The ZFC curve by definition can only be measured with increasing temperature, but it was reproducible when the measurement was repeated after heating above the irreversibility temperature T_i following the temperature sweeps in the FC state.

There is, however, considerably stronger divergence between the ZFC and FC curves of $\chi(T)$ for all the ternary CrCoV alloys, as seen in figure 2. The peak in the ZFC curve corresponds to the Néel transition, as determined from the anomaly in the temperature dependence of the resistivity, neutron diffraction, and the magnetoelastic properties [8, 9]. The Néel temperature thus identified by the position of the peaks in figures 1 and 2 drops progressively with the addition of V, from $T_N = 330$ K in the binary alloy to $T_N = 165$ K in (Cr + 3.2% Co)$_{99}$V$_1$. The irreversibility limiting temperature $T_i(x)$, where the ZFC and FC curves of $\chi(T)$ diverge, drops rather less rapidly than $T_N(x)$ with increasing V content x. The resultant phase diagram is illustrated in figure 3(a), while figure 3(b) shows how the magnitude of the hysteresis at low temperature increases with V content.

There is a change of slope in the FC curve of $\chi(T)$ at the Néel transition for $x = 0.4$ and 1.0% V at the same temperature T_N shown by the peak in the ZFC curve, as seen in figures 2(c) and (d). In the case of the alloys containing $x = 0$, 0.1 and 0.3% V, there is a peak in the FC curve, which occurs at a slightly lower temperature than T_N, as seen in figures 1, 2(a) and (b). The lower-concentration ternary alloys and the binary alloy show similar temperature dependence for the FC and ZFC curves below T_N (figures 1, 2(a) and (b)), while they are rather different for higher concentrations of V (figures 2(c) and (d)). The commensurate SDW phase appears in binary Cr$_{1-y}$Co$_y$ alloys at $y \sim 2\%$ Co [9, 10]. In ternary CrCoV alloys the addition of V is expected to move the system towards the incommensurate SDW phase [5], so that the change in the nature of $\chi(T)$ in the FC curves with increasing V content is probably related to a phase transition from the commensurate to the incommensurate SDW.

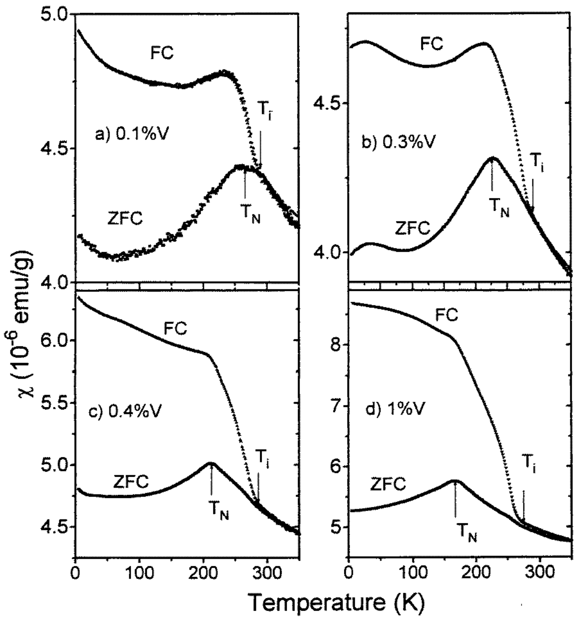

Figure 2. Temperature dependence of the magnetic susceptibility of the ternary alloys (Cr + 3.2% Co)$_{1-x}$V$_x$: $x = 0.1$, 0.3, 0.4 and 1.0% V, measured in a field of 100 Oe after zero-field cooling (ZFC), and in the same field while cooling from a temperature above 400 K and then warming (FC). The Néel temperature T_N, as determined by the peak in the ZFC curve, and the irreversibility limiting temperature T_i, are indicated.

It is clear that the irreversibility in this system is not caused by the formation of a spin glass due to frustration of the moments on the Co atoms: (i) the peak in $\chi(T)$ seen in both the FC and ZFC states is associated with the onset of SDW ordering at the bulk Néel transition rather than a spin-glass freezing transition of the Co moments; (ii) the fact that the irreversibility limiting temperature in a spin glass may be somewhat higher than the freezing temperature for a small cooling field [11], or for clustering of the magnetic impurity, does not explain the behaviour of the present system, in which for $0.1 \leqslant x \leqslant 1.0\%$ V, $T_i(x)$ decreases much more slowly than $T_N(x)$ with increasing x in this range (figure 3(a)); (iii) the relative magnitude of the hysteresis increases linearly with V concentration (figure 3(b)), while the concentration of Co remains constant at 3.2%.

Kostina *et al* [12] predicted the existence of a local spin-density wave associated with non-magnetic impurities in Cr by including electron–impurity interactions in the model for nesting Fermi surfaces used to calculate the SDW properties of the Cr system [5]. Tugushev [13] has reviewed the theoretical work on this problem, which shows that at some temperature T_{loc} above the Néel temperature T_N a local SDW appears at an impurity or other defect, while its nature depends upon whether the long-range order that appears at T_N is a commensurate or incommensurate SDW. In either case the local SDW will carry a local moment that will enhance the susceptibility between T_{loc} and T_N.

The occurrence of C–W paramagnetism in dilute CrV alloys was claimed by Kostina *et al* [12] to correspond to the occurrence of this local SDW phase. There is, however, no evidence in this work (see figure 6 of [14]), nor in later work [15, 16], for a phase transition where the C–W paramagnetism appears, thus defining T_i, at any temperature up to 400 K.

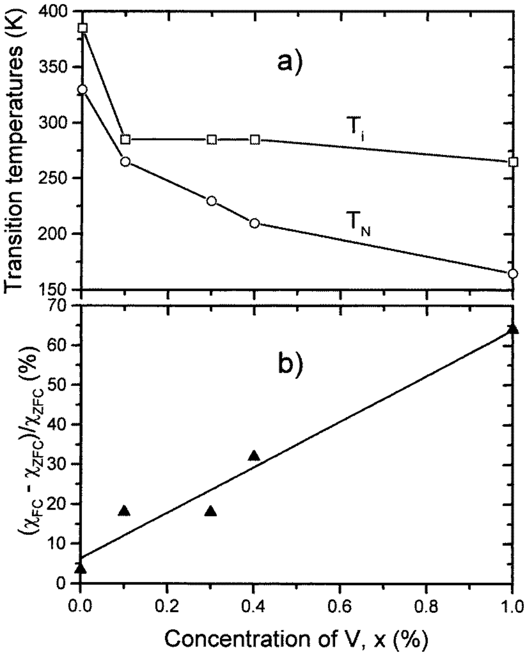

Figure 3. (a) Magnetic phase diagram of the ternary alloy system (Cr + 3.2% Co)$_{1-x}$V$_x$ showing the new phase lying between the Néel temperature T_N (□) and the irreversibility limiting temperature (○). (b) The dependence on V content of the relative magnitude of the hysteresis, $\Delta\chi = (\chi_{FC} - \chi_{ZFC})/\chi_{ZFC}$, at temperature 5 K.

Work is in progress on CrFeV and CrFeMo alloys, where irreversibility effects occur similar to those seen in CrCoV alloys. Binary CrV alloys show only a very small degree of irreversibility, so the effects appear to be associated with the existence of a moment on the Co or Fe atom in the paramagnetic phase.

We believe that the anomaly in the temperature dependence of the susceptibility seen in (Cr + 3.2% Co)$_{1-x}$V$_x$ alloys at the irreversibility limiting temperature T_i corresponds to a new magnetic phase transition, which we tentatively identify with a transition to a local-SDW phase. We encourage further theoretical work on the nature of the local SDW associated with magnetic impurities like Co.

We plan to study the new phase by means of neutron diffraction, anticipating that the irreversibility corresponds to the existence of polarization domains in the local-SDW phase. The new phase transition will also be explored by measuring the temperature dependence in CrCoV alloys of the resistivity, thermal expansion and the elastic constants.

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