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1998 J. Phys.: Condens. Matter 10 4911

(http://iopscience.iop.org/0953-8984/10/22/015)

J. Phys.: Condens. Matter **10** (1998) 4911–4917. Printed in the UK PII: S0953-8984(98)89124-8

The moment of Fe in a $Cr_{1-x}V_x$ host: II. Effect of magnetic field in the spin-density-wave phase

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Received 11 November 1997, in final form 16 March 1998

Abstract. Curie–Weiss fits to the temperature dependence from 5 to 350 K of the susceptibility of Cr + 2.7% Fe doped with up to $x \approx 1\%$ V give different values of the effective moment μ_S for the spin-density-wave (SDW) and μ_P for the paramagnetic phase. For field $H = 1$ T, $\mu_S \approx 0.6\ \mu_P$ in the binary alloy ($x = 0$) and the ratio $\mu_S(x)/\mu_P(x)$ increases as the Néel temperature $T_N(x)$, and therefore the amplitude of the SDW, falls with increasing x. When the field is increased to $H = 5$ T, $\mu_S(x)$ increases towards the full field-independent value of $\mu_P(x)$. It is proposed that interaction with the SDW reduces the Fe moment $\mu_S(x)$, while increasing field tends to restore it to the value $\mu_P(x)$ seen at all fields in the paramagnetic phase.

1. Introduction

Several magnetic impurities carry a magnetic moment in chromium in the paramagnetic phase, as shown by the Curie–Weiss (C–W) temperature dependence of the susceptibility [1]. But only Fe appears to carry a moment also in the spin-density-wave (SDW) phase even at low concentrations, though in general with a smaller C–W coefficient than in the paramagnetic phase [2, 3].

The effect of Fe impurity on the SDW, on the other hand, is rather similar to that of the other ferromagnetic metal Co that carries a moment in the paramagnetic phase. Thus at low impurity concentrations both decrease the Néel temperature T_N at about the same rate, $dT_N/dx = -25$ and -20 K/at.% for $x =$ Fe and Co, respectively (see table 1 in [4]); and both decrease the amplitude of the SDW, but only very slowly (see figure 5(b) in [1]). It is interesting to note that Ni, which does not carry a moment in paramagnetic Cr, decreases T_N very rapidly, with $dT_N/dx = -140$ K/at.% Ni, and also decreases the SDW amplitude much more rapidly than do Fe or Co [1].

In the present paper, we report our finding that the effective Fe moment μ_S obtained from a fit to a C–W law of the temperature dependence of the susceptibility $\chi(T)$ in the spin-density-wave phase alloys in the ternary system $(Cr + 2.7\%\ Fe)_{1-x}V_x$, as well as in binary $Cr_{1-y}Fe_y$ alloys, is substantially reduced below the value μ_P in the paramagnetic phase. In the ternary CrFeV alloy system, doping with V reduces the amplitude of the SDW and thereby increases μ_S until it equals μ_P at $x \gtrsim 1\%$ V (all concentrations are in atomic per cent).

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0953-8984/98/224911+07$19.50

The loss of moment due to interaction with the SDW may also be partially restored in the Cr + 2.7% Fe alloy, and fully restored in the ternary alloys for concentrations $x \gtrsim 0.6\%$ V, by employing a measuring field $H = 5$ T instead of the normal 1 T. Neither the loss of moment in the SDW phase, nor its restoration by a field, is understood theoretically. The explanation of these phenomena may have wider implications for other physical systems, and indeed for the general theory of many-body effects in magnetic systems.

We show in companion paper [4] how the change in the effective Fe moment μ_P in the paramagnetic phase with V doping in $(\mathrm{Cr} + 2.7\%\ \mathrm{Fe})_{1-x}\mathrm{V}_x$ alloys may be described by an environment model, in which the moment of the Fe atom is $\sim 3\ \mu_B$ provided its nearest neighbours are all Cr atoms, but is suppressed completely by a single V neighbour. In the present paper we employ the data from [4] for the effective moment μ_S in the SDW phase in the more dilute alloys ($x \lesssim 1\%$ V) of this system, supplemented by results for binary $\mathrm{Cr}_{1-y}\mathrm{Fe}_y$ alloys and another ternary alloy system $(\mathrm{Cr} + 1.5\%\ \mathrm{Fe})_{1-x}\mathrm{V}_x$, and by high-magnetic-field studies of $\chi(T)$ and the field dependence of the magnetization $M(H)$, to show that:

(1) μ_S is smaller than μ_P in the binary $\mathrm{Cr}_{1-y}\mathrm{Fe}_y$ alloys, a remarkable effect that was first observed by Ishikawa *et al* [2];

(2) μ_S progressively increases towards the more slowly decreasing value μ_P of the effective moment in the paramagnetic phase as the SDW phase is suppressed by adding V;

(3) when the measuring field is increased from $H = 1$ T to 5 T the value of μ_S in Cr + 2.7% Fe obtained from a fit to a C–W law increases from 1.7 μ_B to 2.4 μ_B, while the value $\mu_P \sim 3.0\ \mu_B$ in the paramagnetic phase remains unchanged;

(4) with $H = 5$ T the value of μ_S is restored to the value μ_P for $x \gtrsim 0.6\%$ V, with a single fit to a C–W law straddling the Néel temperature;

(5) the field dependence of the magnetization $M(H)$ in the SDW phase of binary and ternary alloys of Cr containing Fe always exhibits the feature, remarkable for a paramagnet, that $\mathrm{d}M/\mathrm{d}H$ increases with field H. Thus the $M(H)$ curve has positive curvature until, for low temperatures and higher concentrations of Fe, the effect is overwhelmed by the temperature dependence of the magnetic interactions that give negative curvature of $M(H)$ at higher temperatures.

2. Experiment

The measurements in a magnetic field $H = 5$ T were performed with a different SQUID magnetometer MPMS-5S by Quantum Design than those with MPSM-5 in $H = 1$ T reported previously [4]. But the experimental procedures were however identical, and the earlier results were found to be quite reproducible. The C–W plots for four of the samples shown in figure 1 compare the behaviour in the SDW phase below the Néel temperature for $H = 1$ and 5 T. The fitting parameters in the equation expressing the C–W law,

$$\chi(T) = \chi^0_{S/P} + \frac{C_{S/P}}{(T - T^0_{S/P})} \qquad C_{S/P} = \frac{N\mu^2_{S/P}\mu^2_B}{3k_B} \tag{1}$$

N being the number density of Fe atoms (assumed to be single), are given in table 1 for the SDW phase in these four samples at the two values of the field; and in table 1 of [4] at $H = 1$ T for both the SDW and paramagnetic phase in all the samples. In these tables and in (1), the fitting parameters are denoted χ^0_S and χ^0_P; C_S and C_P; and T^0_S and T^0_P, the subscripts S/P corresponding to the SDW and paramagnetic phases, respectively.

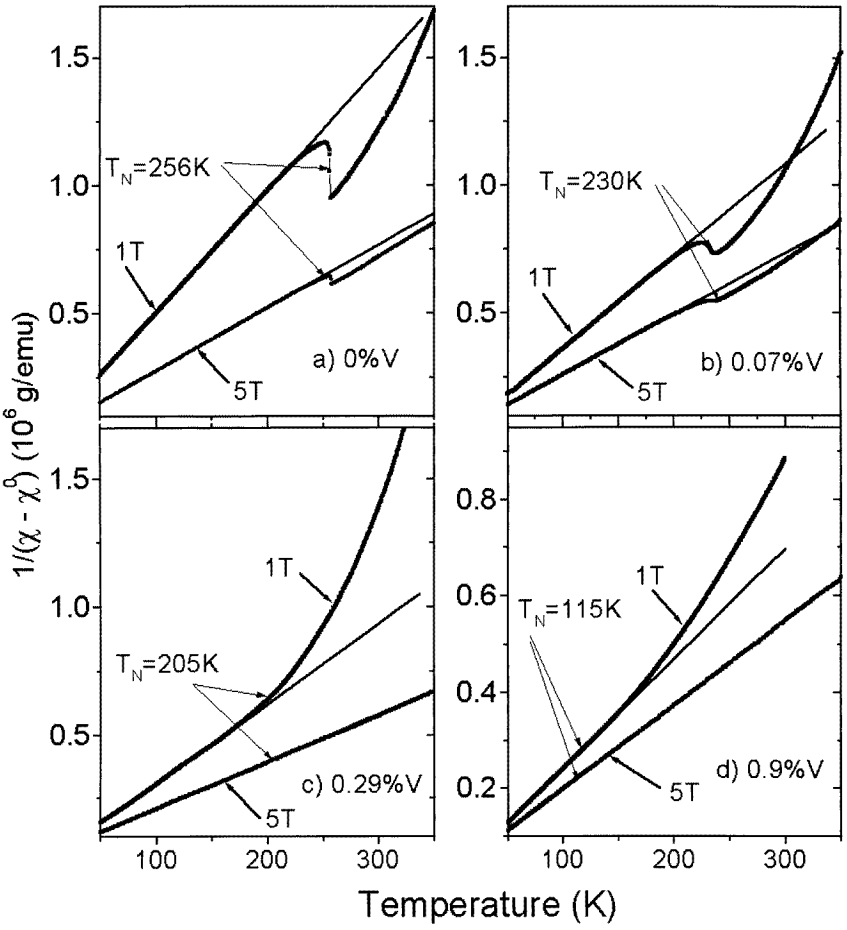

Figure 1. Temperature dependence of the magnetic susceptibility of $(Cr+2.7\%\ Fe)_{1-x}V_x$ alloys measured in field $H = 1$ T and 5 T, as indicated, fitted by the Curie–Weiss law for a range of temperatures below the Néel temperature T_N (except for the $x = 0.9\%$ V sample in panel (d): see text). The fitting parameters are given in table 1.

Table 1. Parameters characterizing the temperature dependence of the susceptibility of $(Cr + 2.7\%\ Fe)_{1-x}V_x$ alloys in the SDW phase, in a fit to a Curie–Weiss law, for measuring fields $H = 1$ T and 5 T.

x (%V)	T_N (K)	T^0_{1T} (K)	T^0_{5T} (K)	χ^0_{1T} (10^{-6} emu g^{-1})	χ^0_{5T} (10^{-6} emu g^{-1})	C_{1T} (10^{-6} K emu g^{-1})	C_{5T} (10^{-6} K emu g^{-1})	μ_{1T} (μ_B)	μ_{5T} (μ_B)
0	256	−5	−13.2	3.88	3.38	205	408	1.71	2.42
0.07	230	0	−10.2	4.00	3.49	276	422	1.99	2.46
0.29	205	2	−12.6	4.05	3.23	319	537	2.14	2.78
0.9	115	−16	−10.1	3.53	3.32	513	515	2.71	2.87

3. Discussion

In figure 2 the dependence of $\mu_P(x)$ on the V concentration is seen to fit the environment model for the paramagnetic phase [4], for both $H = 1$ and 5 T. The behaviour of $\mu_S(x)$

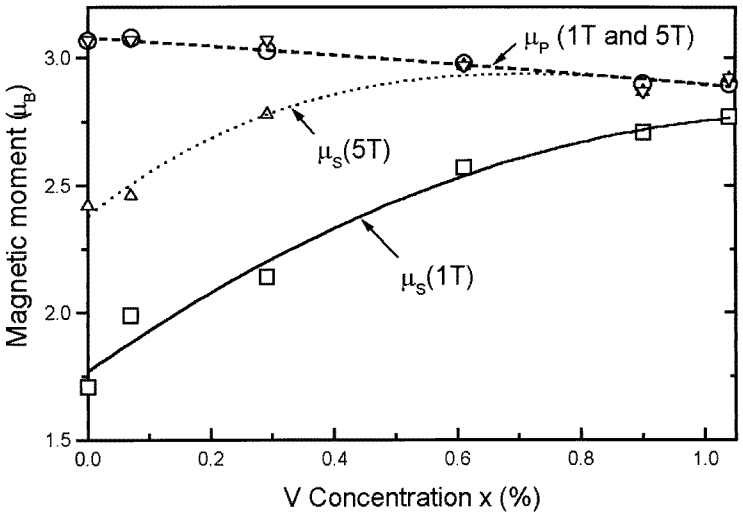

Figure 2. Effective magnetic moment on the Fe atom in $(\mathrm{Cr} + 2.7\%\ \mathrm{Fe})_{1-x}\ \mathrm{V}_x$ in the SDW (μ_S) and paramagnetic (μ_P) phase at $H = 1$ T and 5 T. The dashed line for μ_P at both $H = 1$ and 5 T is in accordance with the environment model.

in the SDW phase concerns us here. For the binary alloy containing $y = 2.7\%$ Fe with $x = 0\%$ V, μ_S is reduced to about 60% of μ_P for $H = 1$ T, but when the measuring field is increased to 5 T the magnitude of μ_S rises to about 80% of μ_P. When V is added, $\mu_S(x)$ for $H = 1$ T rises rapidly towards $\mu_P(x)$ with increasing x as the Néel temperature drops. The loss of moment μ_S in the SDW phase relative to its value μ_P in the paramagnetic phase is substantially restored by the larger measuring field $H = 5$ T, until for $x \gtrsim 0.6\%$ V, $\mu_S = \mu_P$ for $H = 5$ T. The behaviour of the ternary alloy system $(\mathrm{Cr} + 1.5\%\ \mathrm{Fe})_{1-x}\ \mathrm{V}_x$ is similar, with $\mu_S/\mu_P = 0.8$ in the binary alloy $\mathrm{Cr} + 1.5\%$ Fe for $H = 1$ T.

Another way to examine the field dependence of the effective moment in the SDW phase is illustrated in figure 3, where we show the isothermal field dependence of the magnetization M and its derivative $\mathrm{d}M/\mathrm{d}H$ for the binary alloy $\mathrm{Cr} + 2.7\%$ Fe as functions of magnetic field H up to 5 T for temperatures between $T = 2$ and 250 K, i.e., just below the Néel temperature $T_N = 256$ K. The data below 50 K in figure 3 are complementary to those represented in the C–W plots of figure 1(a), which were cut off at this temperature because of the increasingly strong non-linearity of $M(H)$ at lower temperatures. This non-linearity is due to magnetic interactions, which are to be expected at this relatively high concentration of Fe impurity atoms, since even with random distribution only about 80% will be isolated.

The magnetization may be considered to be made up of two components in addition to the single-moment C–W contribution, which is linear in field with a coefficient that varies inversely with temperature. The first is responsible for the approach to saturation of the magnetization at lower temperatures manifested in the decrease of $\mathrm{d}M/\mathrm{d}H$ at higher fields. Pulsed field measurements up to $H = 33$ T at $T = 4.2$ K have shown complete saturation of this component already by 10 T at liquid helium temperatures in $\mathrm{Cr} + 2.4\%$ Fe [5]. The other component has the remarkable characteristic that it causes $\mathrm{d}M/\mathrm{d}H$ to increase with field, as seen at lower fields for all the curves in figure 3. The sum of these two components gives rise to the peak in $\mathrm{d}M/\mathrm{d}H$, which becomes increasingly pronounced and moves to lower fields as the temperature decreases.

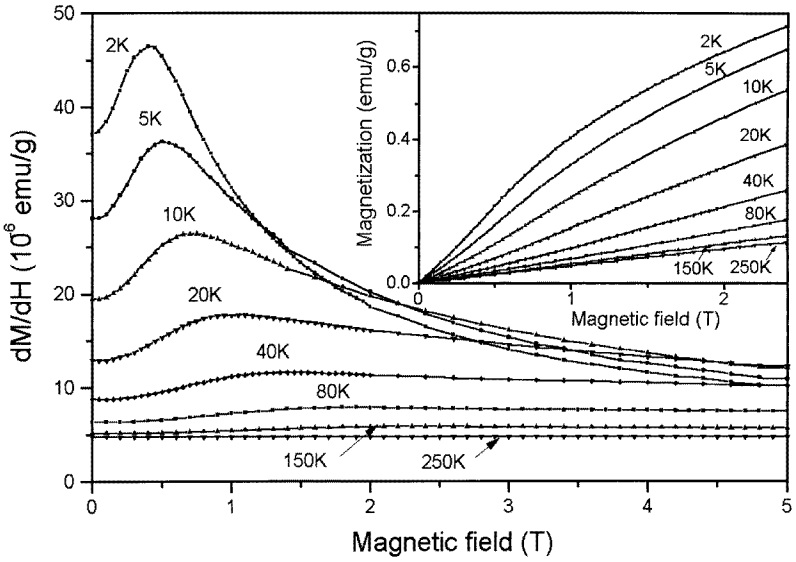

Figure 3. Field dependence of the differential magnetic susceptibility $\mathrm{d}M/\mathrm{d}H$ for the binary alloy Cr + 2.7% Fe in the SDW phase at the temperatures indicated. The corresponding plots for magnetization $M(H)$ are shown in the inset.

To our knowledge this positive curvature of $M(H)$, which in combination with the saturation of $M(H)$ at higher fields gives rise to the point of inflection in $M(H)$, i.e., a maximum in $\mathrm{d}M/\mathrm{d}H$, is a unique feature of Cr alloys that have C–W paramagnetism in the SDW phase. We have observed this behaviour in the other ternary alloy systems based on Cr + 1.5% Fe, as well as in $Cr_{1-y}Mn_y$ alloys, where the Mn atom has a moment in the SDW phase for $x \gtrsim 6.2\%$ Mn [6].

In all our V-doped CrFe alloys the maximum in $\mathrm{d}M/\mathrm{d}H$ is observed in the SDW phase at all temperatures up to the Néel transition. It is absent in the paramagnetic phase, and in particular does not occur at any temperature in the paramagnetic alloys $(\mathrm{Cr}+2.7\%\ \mathrm{Fe})_{1-x}\mathrm{V}_x$ ($x = 5$, 10 and 20% V) and $(\mathrm{Cr}+1.5\%\ \mathrm{Fe})_{1-x}\mathrm{V}_x$ ($x = 3$ and 5% V), which show negative curvature of $M(H)$ at all temperatures.

In the case of Cr+2.7% Fe, which has a strongly first-order Néel transition, as shown by the jump at T_N in the thermal expansion [1], and in the amplitude of the SDW obtained by neutron diffraction [7], the susceptibility also exhibits a singularity, as shown in the inset to figure 4(a). The disappearance of the characteristic peak in $\mathrm{d}M/\mathrm{d}H$ at the Néel temperature $T_N = 256$ K may be seen clearly by comparing the two curves in figure 4(a). The peak in $\mathrm{d}M/\mathrm{d}H$ in this alloy at $T \lesssim T_N$ occurs at this high temperature because it is associated with the coexistence of the local Fe moment and a relatively large-amplitude SDW right up to the Néel transition. In Cr + 1.5% Fe, the Néel transition is quasi-continuous, as shown in the inset to figure 4(b), and $\mathrm{d}M/\mathrm{d}H$ simply changes from an increasing to a decreasing function of H as one increases temperature through $T_N \approx 260$ K, as seen in figure 4(b). This is because of the small amplitude of the SDW close to T_N in this alloy on the one hand, and on the other the reduced effect of magnetic interactions with the lower Fe content.

An interaction between the SDW in Cr and the local moment was postulated by Lomer [8], but in a form that resulted in a decrease of the Curie constant, with a singularity at zero temperature in a model that neglects magnetic interactions between the moments [9].

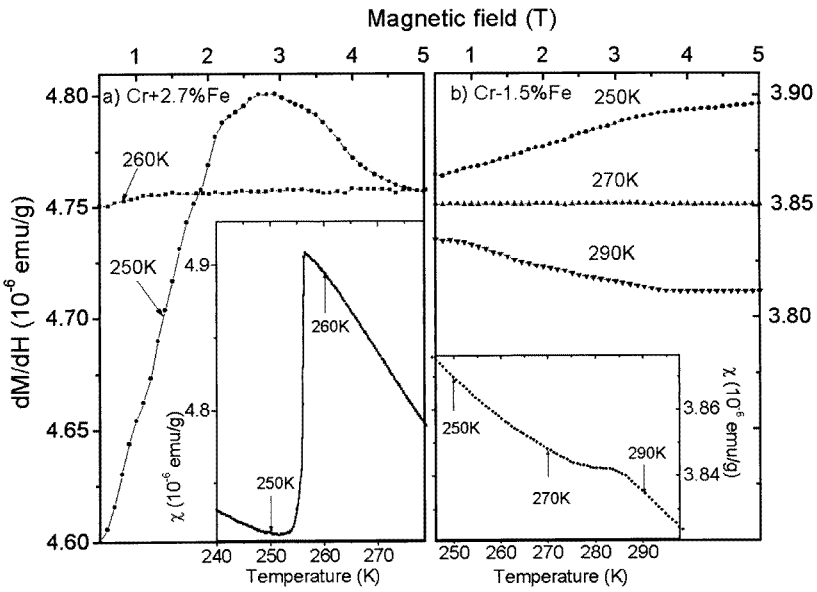

Figure 4. Temperature dependence of the magnetization of CrFe alloys around the Néel temperature; (a) Cr + 2.7% Fe with $T_N = 256$ K; (b) Cr + 1.5% Fe with $T_N \approx 280$ K.

The reduction of the effective moment in the SDW phase that we obtain from a fit to the C–W law indicates however an interaction with the SDW that produces an effect over a wide range of temperatures. We note however that the fit excludes low temperatures from the analysis of the experimental data because of the strong effect of magnetic interactions between the Fe moments in this region. A theory of the interaction between the SDW and the local moment should take into account the temperature dependence of its amplitude. Indeed it might explain the good fit of $\chi(T)$ to the C–W law as resulting from an accidental cancellation between this effect and the temperature dependence of the effect of magnetic interactions between the Fe moments.

The theory should explain also the field dependence of the magnetization associated with the local moment in the SDW phase, which is demonstrated in figure 2 by the increase with increasing field from $H = 1$ T to 5 T of the effective moment in a fit of $\chi(T)$ to the C–W law, and by the positive curvature of $M(H)$ at lower fields as seen in figure 3. The value χ_P^0 of remains essentially constant, as shown by the data points, which are the same within the experimental accuracy for both $H = 1$ T and 5 T, as indicated in figure 2.

The physical mechanism responsible for the field effect may be that the field tends to rotate the polarization of the SDW in the incommensurate phase into a transverse direction, whereas the moment on the Fe atom will rotate it towards the field direction [10], thereby reducing the interaction between the SDW and the moment that reduces the magnitude of μ_S in low field. A moment in the single impurity limit that increases with field has been observed in a quite different physical context in LaB_6 doped with Ce [11]. This is not an SDW system, and the increase is largely a crystal-field effect, with other features due to the Kondo effect [12].

It seems that the local moment on the Fe atom in the SDW phase of CrFe in CrFeV alloys is partially compensated, with the moment being restored as the SDW amplitude is reduced by doping with V [4]. This effect may be related to the Kondo phenomenon,

and there is some evidence that in the latter case also the application of a relatively large magnetic field may tend to restore the moment [11]. It is not clear how these effects are related to the existence of local impurity states in the energy gap of a SDW system [13], whose existence in CrFe alloys (doped with V or Mn to tune the Fermi level) has been demonstrated in other experiments [14, 15].

Acknowledgments

This work was performed with partial support from the Brazilian Agencies CNPq and FAPESP (VYuG and WAO) and by the Natural Sciences and Engineering Research Council, Canada (EF).

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