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The moment of Fe in a $\text{Cr}_{1-x}\text{V}_x$ host: I. The paramagnetic phase

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Abstract. The temperature dependence of the magnetic susceptibility of $(\text{Cr} + 2.7\% \text{Fe})_{1-x}\text{V}_x$ alloys ($x = 0$ to 20% V), measured over the range $5 < T < 400$ K, is found to fit a Curie–Weiss law, with different parameters above and below the Néel temperature in the SDW alloys. The effective moment in the paramagnetic alloys ($x = 5, 10$ and 20% V), and in the paramagnetic phase of the spin-density-wave (SDW) alloys, varies with the V content in accordance with an environment model in which the Fe moment is $3 \mu_B$, provided none of its nearest neighbours is a V atom, but is suppressed completely by a single V neighbour.

1. Introduction

The question of whether or not a magnetic transition metal impurity atom dissolved in a non-magnetic transition metal exhibits a magnetic moment has been investigated both experimentally [1] and theoretically [2] for many years. The absence of a moment on a single Fe atom in a group 5 (V, Nb, Ta) host, and the occurrence of a moment of about two Bohr magnetons in a group 6 (Cr, Mo, W) host, are well known experimental results [3, 4], which have been reproduced in calculations within the framework of the local spin-density approximation [5].

Special attention has been given to the question of whether or not the local moment on a magnetic atom dissolved in a binary alloy system develops continuously with alloy composition, or discontinuously depending on the nearest-neighbour configuration, as suggested by Jaccarino and Walker [6]. They explained the magnetic behaviour of the ternary alloy system $(\text{Mo}_{1-x}\text{Nb}_x) + 1\% \text{Fe}$ (all percentages are atomic per cent) within the framework of a simple ‘environment model’, in which the Fe atom is assumed to have a constant moment of about $2.1 \mu_B$, provided it is surrounded by seven or eight Mo neighbours in the bcc lattice, i.e. does not have more than one Nb neighbour, while an Fe atom having two or more Nb neighbours is ‘demagnetized’, having zero moment. The Jaccarino–Walker hypothesis has been applied to a large number of systems, as documented by Beuerle *et al* [7], with the term ‘environment model’ meaning that the moment on the magnetic atom is determined by the number of nearest-neighbour solute atoms.

In the last decade the problem of local moment formation on impurities in both pure metals and alloy systems has received renewed attention, with precise electronic structure

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calculations being performed that exploit new computing facilities [5,7,8]. Super-cell calculations in the local spin-density approximation [7,8] roughly confirm the environment model for Fe and Co in a $\text{Mo}_{1-x}\text{Nb}_x$ matrix.

We report here an experimental study of temperature dependence of the susceptibility in the ternary alloy system $(\text{Cr} + 2.7\% \text{Fe})_{1-x}\text{V}_x$, $0 < x < 20\%$ V. This system is of special interest because CrFe alloys are the only binary antiferromagnetic alloys of Cr that, even at low concentrations, exhibit Curie–Weiss (C–W) paramagnetism in the spin-density-wave (SDW) phase as well as in the paramagnetic phase (see table V and figure 60 in [9]). We interpret the C–W behaviour that is seen in the SDW phase down to temperatures of about 50 K as corresponding to the moments on single Fe atom impurities substituted into the CrV lattice. Previous studies of the susceptibility in CrFeV alloys are summarized in [10].

The SDW in Cr+2.7% Fe is suppressed by adding a little more than 2% V (see figure 1 in [11]). We find that in the SDW phase the temperature dependence of the susceptibility obeys a C–W law, with a Curie constant that is only about half as big as in the paramagnetic phase, but which increases with V concentration in $(\text{Cr} + 2.7\% \text{Fe})_{1-x}\text{V}_x$ until it is the same as in the paramagnetic phase for $x \gtrsim 1\%$ V. The behaviour of the local moment on the Fe atom in the SDW phase will be considered in another paper [12].

In the paramagnetic phase the effect of adding V is to decrease the Curie constant for that component of the susceptibility of Cr + 2.7% Fe that obeys a C–W law. If the magnitude of this component is assumed to be a measure of the average moment μ_P on each Fe atom, then our results for the effect of V on μ_P are entirely consistent with an environment model in which the Fe atom has the full moment, $\mu_P \approx 3 \mu_B$, unless it has one or more V neighbours, which completely suppress the moment.

2. Experiment

The method of preparing the CrFeV samples, determining their composition, and characterizing them by measuring the temperature dependence of the resistivity and thermal expansion was described previously [11]. The samples for measurement of the magnetization M were cut from the samples used for thermal expansion measurements. $M(H, T)$ was measured with a SQUID magnetometer (Quantum Design), over the temperature range $5 \leq T \leq 400$ K, in magnetic field, $H = 10$ kOe; and in fields up to $H = 55$ kOe for fixed temperatures in the range, $5 \leq T \leq 100$ K.

We define the magnetic susceptibility by the ratio,

$$\chi(T) = M(T)/H. \quad (1)$$

Only the binary Cr + 2.7% Fe and the ternary alloy lightly doped with 0.07% V show an anomaly in $\chi(T)$ at the Néel temperature T_N , which for the other samples was determined from the temperature dependence of the resistivity [11].

3. Results

Figure 1 shows the temperature dependence of $\chi(T)$ for Cr + 2.7% Fe, for two ternary SDW alloys, and for the paramagnetic alloy, $(\text{Cr} + 2.7\% \text{Fe})_{95}\text{V}_5$. The inset shows $\chi(T)$ in more detail around the Néel temperature T_N . The first-order transition for the binary alloy ($x = 0\%$ V) is slightly broadened when $x = 0.07\%$ V is added, as in the case of the thermal expansion $\varepsilon(T)$ (see figure 5 in [11]). The anomaly at T_N disappears completely for $x = 0.29\%$ V in both $\chi(T)$ and $\varepsilon(T)$, though for the latter an anomaly is seen at the

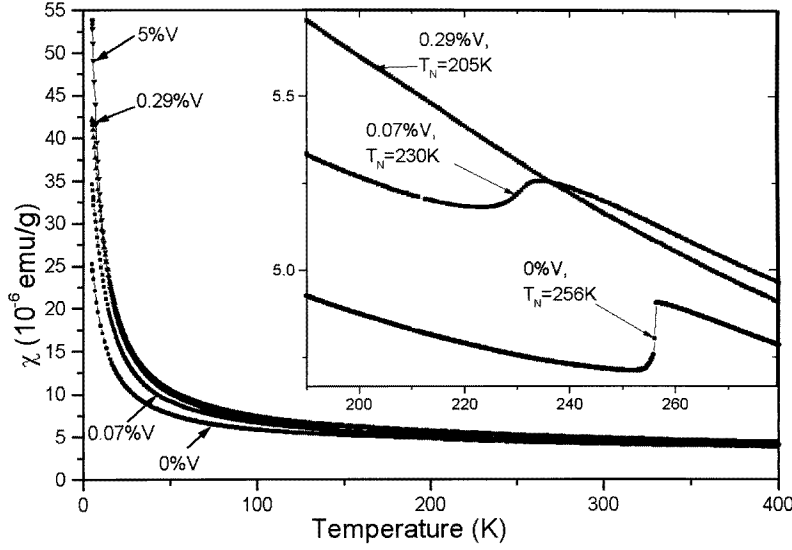


Figure 1. Temperature dependence of the magnetic susceptibility of $(Cr+2.7\% Fe)_{1-x}V_x$ alloys. The curves are labelled with concentration $x\%$ V expressed as percentage. The behaviour around the Néel temperature T_N is shown in more detail in the inset. T_N for 0.29% V-doped alloy is defined from resistivity data [11].

transition temperature, $T_{IC} = 145$ K, from the incommensurate to the commensurate SDW phase.

The plots of the inverse susceptibility at various temperatures for the binary $Cr+2.7\%$ Fe alloy and for the ternary $(Cr+2.7\% Fe)_{1-x}V_x$ alloys that exhibit a different C–W law in the SDW and paramagnetic phases are shown in figure 2. When fitting these data to the C–W expression,

$$\chi(T) = \chi_{S/P}^0 + \frac{C_{S/P}}{(T - T_{S/P}^0)} \quad (2)$$

we employ fitting parameters χ_S^0 and χ_P^0 ; C_S and C_P ; and T_S^0 and T_P^0 for the SDW and paramagnetic phases, respectively. The corresponding values μ_S and μ_P of the effective moment on an Fe atom are obtained from the expression

$$C_{S/P} = \frac{N\mu_{S/P}^2\mu_B^2}{3k_B} \quad (3)$$

where N is the number density of Fe atoms. We note that the breakdown of the C–W law, for $\chi(T)$ below about temperature 50 K, is due to the formation of moments associated with pairs (and clusters) of Fe atoms, which revert to the single-atom moments at higher temperatures due to thermal excitations. The magnetization $M(H)$ becomes linear in field at about the same temperature, as illustrated for the binary alloy in figure 3.

The fit to the C–W law was obtained by varying χ^0 around the optimum value, with C and T^0 as free parameters for each value of χ^0 , so as to obtain the best least-squares correlation coefficient R . The process is illustrated in table 1 for the alloy $Cr+2.7\%$ Fe containing 0.07% V in the spin-density-wave temperature range 65–180 K. In this case we estimate the optimum value of the moment to be $\mu_S = 1.99 \pm 0.02$. A similar procedure

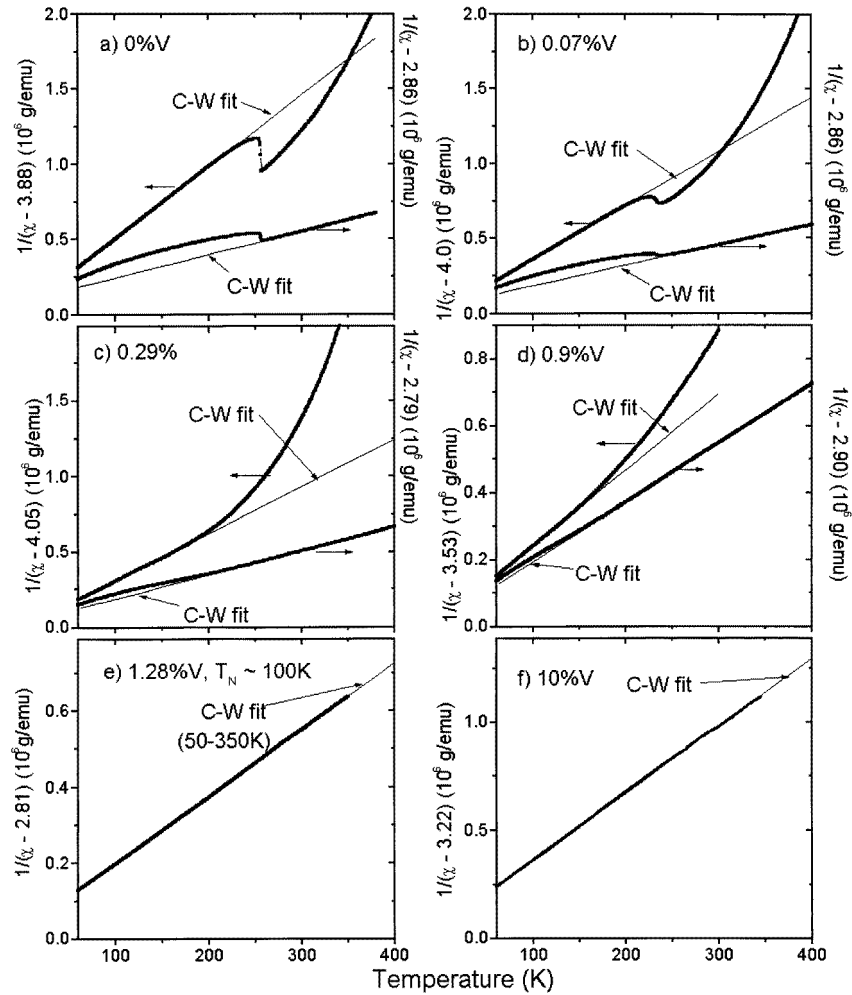


Figure 2. Temperature dependence of the magnetic susceptibility of $(\text{Cr}+2.7\% \text{Fe})_{1-x}\text{V}_x$ alloys fitted to the Curie–Weiss law, with different parameters above and below the Néel temperature T_N , over temperature ranges as indicated in each panel.

was used to obtain μ_P (and μ_S for the SDW phase) for each alloy. The error bars are not shown in the figures however, since this process does not provide a rigorous definition.

The values of the parameters in equations (2) and (3) that give the best fit of the data to a C–W law over the temperature ranges indicated are given in table 2. In most cases the fit is almost perfect in both the SDW and paramagnetic phase, the value of the correlation coefficient typically being, $R = 0.999\,95\text{--}0.999\,995$.

The effective magnetic moments $\mu_{S/P}$ obtained from the fitting parameters for the C–W plots tabulated in table 2 are plotted versus V concentration in figure 4. For the lower-V-concentration SDW alloys, and for the binary CrFe alloy, μ_S is distinctly different from μ_P . But for the two alloys containing 1.28 and 2.14% V, the Néel temperature T_N is so low (though the precise value is not known) that the range in the SDW phase between T_N and the temperature at which deviations from the C–W law begin is too small to make a

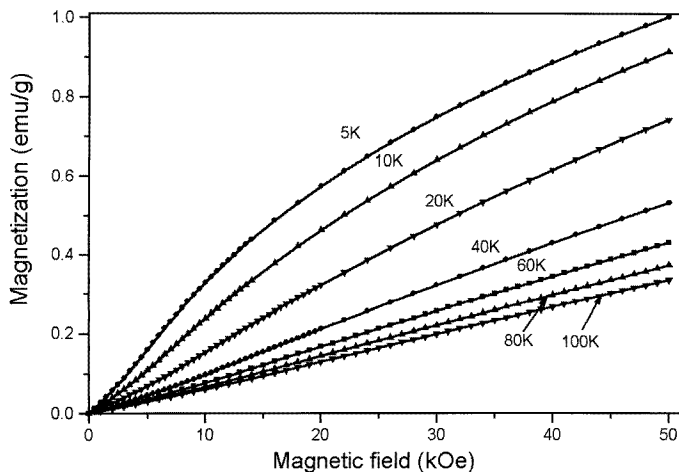


Figure 3. Field dependence of the magnetization $M(H)$ of the binary alloy Cr + 2.7% Fe at various temperatures, as indicated for each curve.

Table 1. Parameters in fits to a Curie–Weiss law for the temperature dependence of the susceptibility in the SDW phase of the alloy containing $x = 0.07\%$ V dissolved in Cr + 2.7% Fe, showing the variation of the least-squares correlation coefficient R as χ^0 is varied around the value 4.00 that gives the best fit.

χ_S (10^{-6} emu g^{-1})	C_S (10^{-6} K emu g^{-1})	T_S^0 (K)	μ_S (μ_B)	R
3.90	302	-5	2.08	0.99995
3.92	297	-4	2.06	0.99997
3.94	291	-3	2.04	0.99998
3.96	286	-2	2.03	0.99999
3.98	282	1	2.01	0.99999
4.00	276	0	1.99	> 0.999995
4.02	272	1	1.97	0.99999
4.04	267	2	1.96	0.99998
4.06	261	3	1.94	0.99997
4.08	257	4	1.92	0.99995
4.10	252	5	1.90	0.99992

separate low-temperature fit to equation (2). Thus only a single point, which corresponds to the value of both μ_P and μ_S , can be obtained from the linear fit in figure 2(e) (and a similar plot for $x = 2.14\%$ V), as plotted in figure 4.

4. Discussion

We shall discuss first the decrease in the effective moment of the Fe atom in $(Cr + 2.7\% Fe)_{1-x}V_x$ as the concentration x of V increases. The behaviour of a similar ternary system, $Mo_{1-x}Nb_x + 1\%$ Fe, appears to fit an environment model in which the moment on an Fe atom changes discontinuously for the full value to zero when the number of Nb nearest neighbours exceeds a critical value [6, 13].

In fact, Jaccarino and Walker [6] erroneously plotted (μ_P/μ_0) , rather than the square of the moment ratio (see (4) and (5) below), versus the concentration x of Nb, and their

Table 2. Parameters characterizing the temperature dependence of the susceptibility of $(\text{Cr} + 2.7\% \text{Fe})_{1-x}\text{V}_x$ alloys in a fit to a Curie–Weiss law.

x (% V)	T_N (K)	T_S^0 (K)	T_P^0 (K)	χ_S^0 (10^{-6} emu g $^{-1}$)	χ_P^0 (10^{-6} emu g $^{-1}$)	C_S (10^{-6} K emu g $^{-1}$)	C_P (10^{-6} K emu g $^{-1}$)	μ_S (μ_B)	μ_P (μ_B)	Fitting T range (K)	
										SDW	Param.
0	256	−5	−61	3.88	2.86	205	656	1.71	3.07	50–225	260–380
0.07	230	0	−17	4.00	2.86	276	662	1.99	3.08	65–180	260–400
0.29	205	2	−22	4.05	2.79	319	640	2.14	3.03	65–180	240–370
0.61	155	−7	−24	3.50	2.90	461	621	2.57	2.98	50–170	185–360
0.9	115	−16	−16	3.53	2.90	513	588	2.71	2.90	50–100	170–400
1.04	110	−4	−8	3.44	2.87	535	585	2.77	2.90	50–100	170–400
1.28	× ^a		−11		2.81		563		2.84		150–350
2.14	×		−14		3.20		549		2.80		50–400
5			−12		3.00		448		2.53		50–300
10			−17		3.22		319		2.13		50–340
20			−2		3.28		55		0.89		50–330

^a This alloy has an SDW phase, but the value of T_N cannot be determined accurately from the temperature dependence of the resistivity as it is for the other SDW alloys [9]. In the case of the alloy containing 1.28% V, the fitting temperature range extends from the paramagnetic phase to well below the Néel temperature, as shown in figure 2(e). These mean that the effective moment is the same in both phases.

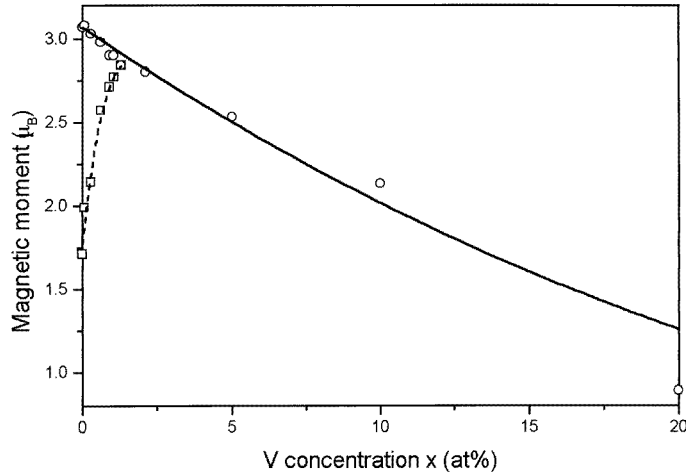


Figure 4. Effective magnetic moment on the Fe atom in $(\text{Cr} + 2.7\% \text{Fe})_{1-x}\text{V}_x$ alloys as a function of V concentration: \circ — μ_P for paramagnetic alloys and for the paramagnetic phase of SDW alloys; \square — μ_S for the SDW phase. The curve fit to the paramagnetic data points corresponds to the probability function $P_8(x)$ defined in (5) and shown also in figure 5. The dash curve through the $\mu_S(x)$ data points is a guide to the eye.

fit of the data of Clogston *et al* [3] to the probability function $P_7(x)$ defined in (5) below was considered by Nagasawa and Sakai [13] to be fortuitous. The latter workers measured the effective moment on the Fe atom in five alloys of $\text{Mo}_{1-x}\text{Nb}_x$ ($x = 0.4, 0.3, 0.2, 0.1$ and 0) with concentrations of Fe impurity ranging from 0.2 to 1.0%, and for the first three obtained values considerably different from those of Clogston *et al*, which they attribute to error in the analysis of the Fe concentration in the earlier work; their own data fitted very well the $P_7(x)$ probability function.

We analyse our data for the paramagnetic alloys according to the environment model by considering that, for a given concentration x of V in a $(\text{Cr} + 2.7\% \text{Fe})_{1-x}\text{V}_x$ alloy, the magnetization is due to N_{mag} Fe atoms that have the full moment, $\mu_0 \approx 3 \mu_B$, corresponding to $x = 0$, i.e. the value in the paramagnetic phase of the binary Cr + 2.7% Fe alloy. The value $\mu_P(x)$ of the paramagnetic moment in table 2 is calculated from the experimental value of the Curie constant C , with the assumption that *all* the Fe atoms have the same effective moment, so that we can write

$$N\mu_P^2(x) = N_{mag}(x)\mu_0^2. \quad (4)$$

We now compare in figure 5 the ratio, $(\mu_P(x)/\mu_0)^2$, which from (4) is seen to be a measure of $N_{mag}(x)/N$, with the functions

$$P_n(x) = \sum_{r=n}^8 \frac{8!}{r!(8-r)!} (1-x)^r x^{8-r} \quad (5)$$

which expresses the probability, for random distribution in a binary $\text{Cr}_{1-x}\text{V}_x$ alloy, that a single Fe atom substituted for a Cr atom has $(8-n)$ or fewer V neighbours. The fit to the curve $P_8(x)$ is quite good, which according to the environment model means that a single V nearest neighbour is enough to suppress the moment of an Fe atom substituted in the Cr lattice.

It is interesting to compare this with the behaviour of an Fe impurity atom in the $\text{Mo}_{1-x}\text{Nb}_x$ matrix and of Co in $\text{Cr}_{1-x}\text{V}_x$. The good fit to the $P_7(x)$ probability function

in the case of Fe impurity in $\text{Mo}_{1-x}\text{Nb}_x$ alloys [13] means that two Nb nearest neighbours are needed to suppress the moment on the Fe atom. The system $\text{Cr}_{1-x}\text{V}_x + 1\%$ Co has also been measured [14], with results that are ambiguous, since the data points lie between the curves $P_7(x)$ and $P_8(x)$ in a plot like figure 5. Nevertheless, the environment model seems to be well established, with a study of the Mossbauer effect in $\text{Mo}_{0.8}\text{Nb}_{0.2}$ showing the coexistence of Fe atoms carrying zero moment and a moment of about $2 \mu_B$ [15]; and a similar result is obtained for NMR measurements of Co in $\text{Cr}_{1-x}\text{V}_x$ alloys [16]. It should be noted that, with 2.7% Fe in each alloy, the probability of an Fe atom having one other nearest-neighbour Fe atom is quite high, about 20%. The assumption of the environment model is that the moment on each Fe atom is nevertheless determined solely by the number of V nearest neighbours. Thus a pair of neighbouring Fe atoms is assumed not to develop a pair moment, as was postulated in other work [10].

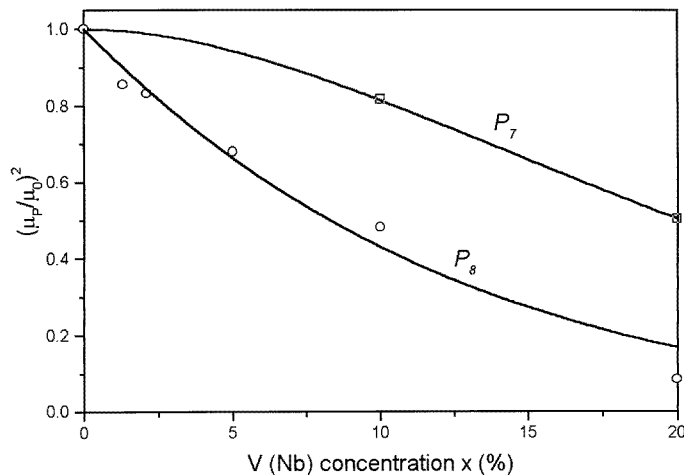


Figure 5. Probability functions $P_n(x)$ ($n = 7, 8$), as defined in (5), showing the probability, for random distribution, that an Fe atom substituted for a Cr atom in $\text{Cr}_{1-x}\text{V}_x$, or for an Mo atom in $\text{Mo}_{1-x}\text{Nb}_x$, has $(8 - n)$ or fewer V (Nb) atoms. The data points (○) for the $\text{Cr}_{1-x}\text{V}_x$ host are seen to fit the curve $P_8(x)$, whereas those (□) for the $\text{Mo}_{1-x}\text{Nb}_x$ host [13] fit $P_7(x)$.

The effective moment μ_S on the Fe impurity in the SDW phase increases progressively with V concentration as it approaches the relatively slowly changing value μ_P at low concentrations of V, as shown in figure 4. This effect is discussed in another paper [12].

5. Conclusion

In the paramagnetic phase of $\text{Cr}_{1-x}\text{V}_x$ the environment model appears to explain nicely the rapid decrease of the average moment μ_P on an Fe impurity atom as V is alloyed into Cr. The fact that a single V nearest neighbour suppresses the moment on the Fe atom is a strikingly simple result. The contrast with $\text{Mo}_{1-x}\text{Nb}_x$, in which two Nb neighbours are needed to suppress the Fe moment, is interesting, and the two systems should be calculated to test the validity of the theory used.

Acknowledgments

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