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1992 J. Phys.: Condens. Matter 4 923

(http://iopscience.iop.org/0953-8984/4/3/032)

J. Phys.: Condens. Matter 4 (1992) 923–928. Printed in the UK

COMMENT

Suppression of spin fluctuations in Cr by doping with V

Eric Fawcett

Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7

Received 17 June 1991, in final form 29 August 1991

Abstract. Spin fluctuations in the paramagnetic phase of the spin-density-wave antiferromagnet Cr are greatly reduced by doping with V, as observed directly by inelastic neutron scattering, and indirectly by the effect on the nuclear magnetic relaxation time, the nature of the Néel transition, the electrical resistivity, the thermal expansion and bulk modulus, and the magnetic susceptibility. The collation here of these previously reported experimental results shows their significance, while the effect of V doping on the magnetoelastic properties of Cr is represented directly, rather than through the change in the magnetic Grüneisen parameter, as previously.

The marked effect of V impurities on the spin fluctuations in the paramagnetic phase of the spin-density-wave (SDW) antiferromagnet Cr was first noticed by Kontani and Masuda [1]. They measured the nuclear spin–lattice relaxation time T_1 of ^{53}Cr in pure Cr and in an alloy, Cr + 2 at.% V, and found that in both cases its dependence on temperature (T) was of the form predicted by the self-consistent renormalization theory for spin fluctuations [2]

$$1/T_1T = c + CT/(T - T_N)^{1/2} \tag{1}$$

T_N being the Néel temperature. The first term is the Korringa relaxation rate, which is found as expected to be approximately the same in the dilute alloy as in the pure metal. The second term, however, shows a decrease in the coefficient C by almost an order of magnitude when the V impurity is introduced.

It is claimed that an analysis [3], including spin fluctuations [2, 4], for the canonical model [5, 6] of nesting electron and hole octahedra with a reservoir, accounts for the progressive decrease [7] of the coefficient C for magnetic resonance of the ^{51}V nucleus with increasing concentration of V. The change in C for the ^{53}Cr nucleus between pure Cr and Cr + 2 at.% V is, however, rather larger than expected. Kontani and Masuda [1] suggest that the remarkably large contribution of spin fluctuations to the nuclear magnetic relaxation in pure Cr follows from its proximity to the incommensurate–commensurate SDW phase transition [5].

In this paper, the effect of V doping on the nuclear spin relaxation is collated with subsequent experiments by the author and coworkers, using inelastic neutron scattering, which show directly similar effects of V on the spin fluctuations in the paramagnetic phase of Cr, and on the magnetoelastic properties, thermal expansion and bulk modulus, these being dramatically affected by V doping. Experimental data on the temperature dependence in the paramagnetic phase of the electrical resistivity

0953-8984/92/030923+06$03.50

and the magnetic susceptibility, which have gone unnoticed in the literature for some years, are also related to the effects on other properties.

Direct evidence from inelastic neutron scattering for the suppression of spin fluctuations in the paramagnetic phase was first obtained by Fawcett *et al* [8] for a Cr + 0.5 at.% V sample. A detailed study by Noakes *et al* [9,10] compared the inelastic neutron scattering around the Néel temperature in Cr + 0.2 at.% V with that in pure Cr, with the results given in table 1 [11].

Table 1. Parameters of the Sato–Maki model for Cr and Cr + 0.2 at.% V at the same reduced temperature, $t = 1.027(2)$. a: Susceptibility for incommensurate peaks at low energy. b: Incommensurability parameter, $\delta = 1 - Q$. c: Energy difference between commensurate and incommensurate fluctuations. d: Inverse correlation length scaled by temperature factor.

	$10^3\chi^0/r^2$ (μ_B^2 Å^{-2}/atom)	$10^{-3}A^2$ (THz Å^2)	a $(10^6\chi^0/r^2A^2\kappa^4)/(t-1)^2$ (μ_B^2 THz^{-1}/atom)	b $10^2\delta$ (Å^{-1})	c $A^2\delta^2/4$ (THz)	d $10\kappa/(t-1)^{1/2}$ (Å^{-1})
Cr+0.2 at.% V	0.59	0.96	3.4	9.4	2.14	6.5
Cr	1.21	0.80	13.4	7.8	1.22	5.8
Ratio			0.26	1.2	1.75	1.1

Although the nature of the Néel transition is quite different in the two samples, being first-order in pure Cr and continuous in the dilute alloy, as illustrated in figure 1 of [10], the inelastic scattering in the paramagnetic phase in both cases may be described well by a form of the dynamic susceptibility for an incommensurate itinerant antiferromagnet postulated by Sato and Maki [12]

$$\chi(Q,\omega) = (\chi^0/r^2)A^2/\{A^4[\kappa^2 + R(q)]^2 + \omega^2\} \tag{2}$$

where χ^0 is the coefficient of the Curie-law susceptibility in the non-interacting limit, r is the length scale of the magnetic interaction, A is the 'magnetic stiffness', and κ is the inverse correlation length. The self-energy function $R(q)$ may be written

$$R(q) = (1/4\delta^2)[(|q|^2 - \delta^2)^2 + 4(q_x^2q_y^2 + q_x^2q_z^2 + q_y^2q_z^2)] \tag{3}$$

which is the simplest polynomial of fourth order in $q = Q-(1,0,0)$ that conveniently represents the sixfold symmetry exhibited by the satellites around the commensurate position, $Q = (1,0,0)$.

The decrease of intensity at the incommensurate peak by nearly a factor four (column (a) of table 1) is quite remarkable at such a low level of V doping. The incommensurability parameter δ increases by only 20% in the alloy (column (b) of table 1), but this small shift away from commensurability is believed [1,13] to be a significant factor in causing the sharp decrease of intensity of the spin fluctuations.

Indirect evidence for the suppression of spin fluctuations by V impurities is provided by the fact, pointed out above, that the Néel transition, which is clearly first order, though weak, in pure Cr, becomes continuous when as little as 0.2 at.% V is introduced [10,14]. The microscopic origin of the weak first-order Néel transition in pure Cr has not yet been explained. A renormalization-group analysis shows, however, that the Ginzburg–Landau–Wilson Hamiltonian has no stable fixed points

[15], which suggests that fluctuations occurring as the transition is approached drive it first order. Fawcett *et al* [16], who first discovered the effect of V doping on the transition by means of thermal expansion measurements, suggested that the random fields associated with the impurities reduce the local symmetry and thus restore a stable fixed point at which the continuous transition occurs.

A more likely explanation for the weak first-order transition was proposed by Young and Sokoloff [17], who found that for some values of the parameters the canonical model [5,6] gives a first-order transition connected with the existence of relatively strong SDW harmonics. The fact that the third-harmonic content of the SDW decreases rapidly with V doping [18] (and conversely increases with Mn doping as the system moves *towards* commensurability) supports this idea. The intensity of the spin fluctuations according to these results is related to the amplitude of the SDW harmonics.

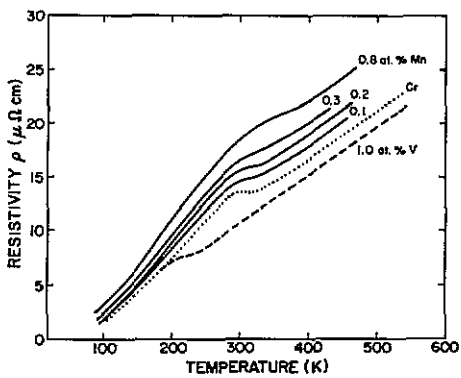

Figure 1. Temperature dependence of the electrical resistivity of pure Cr, Cr + 1.0 at.% V (after Vedernikov [19], Trego and Mackintosh [20]) and dilute alloys of Mn with Cr (after Yakhmi *et al* [21]).

The electrical resistivity in the paramagnetic phase decreases by about 10% over several hundred degrees above the Néel temperature when Cr is doped with 1.0 at.% V, as illustrated in figure 1. This unusual behaviour provides further indirect evidence for the suppression of spin fluctuations, which by analogy with the effects observed in weak itinerant ferromagnets [22] might be expected to contribute to the resistivity in Cr. The decrease of electrical resistivity on doping with V contrasts with the normal increase due to impurity scattering that is seen in figure 1 as Mn is introduced into Cr.

The magnetic contributions $\Delta\beta$ to the thermal expansion and ΔB to the bulk modulus, were obtained for Cr [23] and antiferromagnetic Cr alloys [24] from the difference between the measured values of β and B and those for the paramagnetic alloy Cr + 5 at.% V. The resultant plots of $\Delta\beta(t)$ and $\Delta B(t)$ in the paramagnetic phase, as functions of reduced temperature, $t = T/T_N$, are shown in figures 2 and 3 for pure Cr and for two dilute alloys, with V having almost the same composition in each. There is clearly a dramatic reduction in both $\Delta\beta(t)$ and $\Delta B(t)$ induced by the V impurity, which may be quantified by plotting the values for the two samples measured at the same reduced temperature t, as shown in the upper right-hand parts of figures 2 and 3. The ratio, $\Delta\beta(\mathrm{Cr})/\Delta\beta(\mathrm{Cr} + 0.5\ \text{at.\% V})$, has the value 3.5 for $t > 1.15$. In the case of the bulk modulus, the $B(t)$ curve for Cr + 0.67 at.% V is

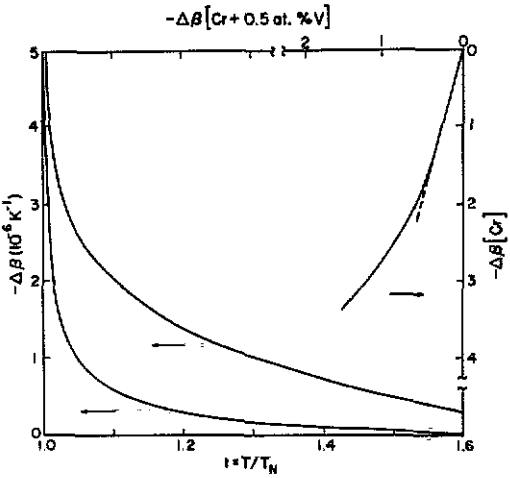

Figure 2. Temperature dependence of the magnetic contribution $\Delta\beta$ to the thermal expansivity β for pure Cr and Cr + 0.5 at.% V. The continuous curves through the data points of White *et al* [27] are taken from Fawcett [25], and the plot of $\Delta\beta$(Cr) against $\Delta\beta$(Cr + 0.5 at.% V) is made to go through the origin by shifting down the curve of $\Delta\beta$ against t for pure Cr.

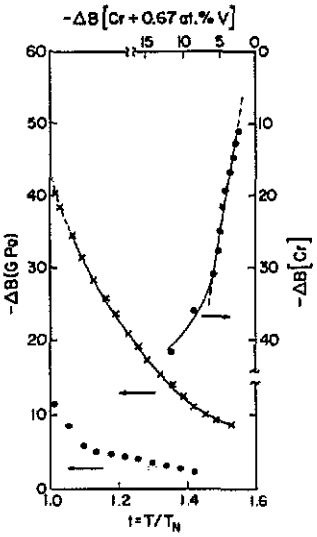

Figure 3. Temperature dependence of the magnetic contribution ΔB to the bulk modulus B for pure Cr and Cr + 0.67 at.% V (after de Camargo and Brotzen [26]).

shifted down by about 10% relative to the curve for pure Cr [26], so that we must evaluate the slope of the plot of ΔB(Cr)/ΔB(Cr + 0.67 at.% V), obtaining a value of seven for the ratio for $t > 1.10$.

It should be noted that the previous report [25] did not bring out so clearly as in figures 2 and 3 the dramatic changes in the thermal expansion β and the bulk modulus B produced by V doping. The representation by the change in the magnetic Grüneisen parameter [25] is succinct, but less revealing of the details of the effects on the magnetoelastic properties than the plots given here, which distinguish clearly the effects on the temperature dependence of β and B. Thus the magnetic Grüneisen parameter decreases by a factor of about two with 0.5/0.67 at.% V doping (a closer examination of the experimental data having revised the earlier estimate [25] that it is reduced by a factor of three so that, in the notation of [25], $\Gamma_+^{II} = -80$ instead of -55 for Cr + 0.5/0.67 at.% V), while the temperature dependence of the magnetic contributions to $\Delta\beta$ and ΔB decrease by factors of 3.5 and 7, respectively, as shown in figures 2 and 3.

The reason is not understood for this strong decrease in the magnetic contributions to the magnetoelastic properties when Cr is doped with V. This remarkable behaviour may well result from a rapid decrease with V doping in the amplitude of the second harmonic of the SDW (i.e. the charge-density wave (CDW) and the associated periodic lattice distortion) that is expected to accompany the observed decrease in the third harmonic [18]. The effect of V doping on the amplitude of the CDW in Cr is at present being investigated [28].

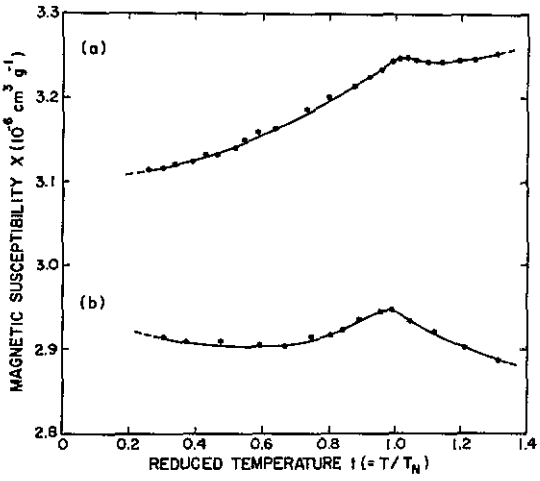

Figure 4. Temperature dependence of the magnetic susceptibility χ of : (*a*) pure Cr; and (*b*) Cr + 0.58 at.% V (after Kondorskii *et al* [29]).

Finally, we note that the temperature dependence of the magnetic susceptibility in the paramagnetic phase is changed qualitatively by the introduction of V [29]. In figure 4 the roughly constant susceptibility of the paramagnetic phase of pure Cr becomes Curie–Weiss like in the dilute alloy. Again the reason for this change is not understood, but it seems likely that it is related to the change in the other physical properties through a common cause.

In summary: (i) changes in both the inelastic neutron scattering and the nuclear relaxation rate provde stong evidence that dilute V impurity suppresses the spin fluctuations in Cr; (ii) the resistivity in the paramagnetic phase decreases with V doping and the Néel transition becomes continuous, both being effects one might expect from the suppression of spin fluctuations; (iii) the magnetoelastic anomalies in the paramagnetic phase are strongly reduced, and the magnetic susceptibility develops a Curie–Weiss temperature dependence.

It would be most desirable to determine the effect of other impurities on the spin fluctuations in Cr by measuring the inelastic neutron scattering, and at the same time to measure the changes in the various physical properties. The data available at present to provide guidance are only fragmentary. Thus Nb (and perhaps Ta [30]) as well as Ti [31] produce a decrease of resistivity in the paramagnetic phase when dissolved in Cr. The magnetic Grüneisen parameter in the paramagnetic phase of $Cr_{1-x}Mo_x$ alloys [24] decreases as in the V alloys, but much less rapidly, by only a factor 3 for $x \simeq 8$ at.%, while Cr + 0.4 at.% Mo appears to have a continuous Néel transition [16]. Both Fe and Co give a Curie–Weiss susceptibility in the paramagnetic phase of dilute alloys with Cr, but this is believed to be due to local moments on these magnetic atoms.

A theoretical treatment of this problem is needed. There has been a great deal of interest in comparing the predictions of self-consistent renormalization theory for weak itinerant ferromagnets with experiment [22, 32], but much less attention has been given to the properties of itinerant electron antiferromagnets in the paramagnetic phase.

Endorsement

It is the author's wish that no agency should ever derive military benefit from the publication of this paper. Authors who cite this work in support of their own are requested to qualify similarly the availability of their results.

Acknowledgment

This work was supported by the Natural Sciences and Engineering Research Council of Canada. The author has benefited from discussions with Dr S H Liu.

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