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1990 J. Phys.: Condens. Matter 2 8779

(http://iopscience.iop.org/0953-8984/2/44/007)

J. Phys.: Condens. Matter **2** (1990) 8779–8790. Printed in the UK

Volume magnetostriction and pressure dependence of the Curie point and spontaneous magnetization of weakly ferromagnetic $Y(Co_{1-x}Al_x)_2$

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Received 12 March 1990, in final form 13 August 1990

Abstract. The pressure dependence of the Curie point of weakly ferromagnetic $Y(Co_{1-x}Al_x)_2$ alloys for $x \sim 0.15$ has been measured at pressures up to 8 kbar. The pressure dependence of the spontaneous magnetization per unit mass at 0 K (σ_0) has been deduced for the same alloys from measurements of the forced volume magnetostriction in fields up to 12 T. It was found that $(\partial \ln T_c/\partial \ln V) = (\partial \ln \sigma_0/\partial P) = 120 \pm 17$, one of the largest values known for a ferromagnetic material. The critical pressure for the collapse of ferromagnetism is $\sim 9 \pm 1$ kbar for $0.14 < x < 0.18$, while the chemical pressure exerted by the Al in $Y(Co_{0.85}Al_{0.15})_2$ is ~ -40 kbar, so weak ferromagnetism would not be possible in this Y–Co–Al system if the material was clamped to the volume of YCo_2.

1. Introduction

The cubic Laves compounds RM_2, where R is a rare earth or Y and M either Fe or Co, have been studied in great detail as examples of metallic magnetism. The series of Y–Fe compounds, including YFe_2, are ferrimagnetic with a moment on the Y site $\sim -0.4\,\mu_B$, (Armitage *et al* 1986, 1989) while YCo_2 is only an enhanced Pauli paramagnet. The density of states function for YFe_2 is quite similar to that of YCo_2, Yamada (1988), but in YCo_2 the fermi level does not lie low enough in energy for the Stoner criterion for ferromagnetism to be satisfied. The addition of Al to YFe_2 reduces the magnetization but Yoshimura and Nakamura (1985) found that $Y(Co_{1-x}Al_x)_2$ becomes a weak itinerant ferromagnet for $x \sim 0.15$. The effect of Al substitution is to change the density of d electrons at the fermi level and to expand the lattice of YCo_2 and one purpose of the present high pressure experiments was to isolate the two contributions to the phase transition.

The theory of weak itinerant ferromagnetism has been intensively studied but the number of materials which satisfy all the theoretical criteria is quite small, e.g. Wohlfarth (1981). While Ni_3Al and $ZrZn_2$ appear to be well described by the spin fluctuation theory in the form developed by Lonzarich and Taillefer (1985) it has recently been confirmed (Grew *et al* 1989) that both the magnetization and Curie point of Sc_3In increase with pressure up to at least 31 kbar while the theory predicts a rapid decrease of both quantities with pressure, Wohlfarth (1981).

Bulk magnetization data and NMR experiments on $Y(Co_{1-x}Al_x)_2$ for $x \sim 0.15$, have been successfully analysed in terms of the spin fluctuation theory of weak itinerant ferromagnetism and we now show that

$$\partial \ln T_c/\partial \ln V = \partial \ln \sigma_{000}/\partial \ln V = 120 \pm 17$$

where σ_{THP} is the magnetization per unit mass at temperature T, magnetic field H and pressure P, a very large value but similar to that found for $ZrZn_2$ (Fawcett and Meincke 1970).

In summary it is concluded that the $Y(Co_{1-x}Al_x)_2$ system exhibits weak itinerant ferromagnetism which is extremely sensitive to pressure, and that, despite the random nature of the alloy, carefully prepared samples exhibit at least as sharp a magnetic transition as well known weak ferromagnets such as $ZrZn_2$. The lattice expansion between YCo_2 and $Y(Co_{0.85}Al_{0.15})_2$ is equivalent to a chemical pressure of ~ -40 kbar, but only 9 kbar is required to reduce the ferromagnetic transition temperature to 0 K so it would be impossible for this Y–Co–Al system to order if clamped to the volume of YCo_2.

The theory of the magnetism of YCo_2 is discussed in section 2, a summary of earlier experimental work is given in section 3, the experimental details of the present experiments in section 4, and a discussion of the results in section 5. Some of the magnetostriction data has already been published (Riedi *et al* 1988).

2. Theory

2.1. Weak itinerant ferromagnetism

The necessary condition for a metal to exhibit magnetic order at 0 K may be written in terms of the Stoner–Wohlfarth theory as $\bar{I} > 1$, where $\bar{I} = ID(\varepsilon_F)$, with I the intramolecular exchange parameter and $D(\varepsilon_F)$ the density of states at the Fermi level for one spin direction, e.g. Wohlfarth (1981). In a weak ferromagnet the inequality is only just satisfied, $(\bar{I} - 1) < 10^{-2}$ for the Y–Co–Al system, so the T_c and spontaneous magnetization at 0 K are both small.

The application of pressure to a weak itinerant ferromagnet leads to a broadening of the electron bands and a decrease in the Curie point and spontaneous magnetization at 0 K. Wohlfarth (1981) showed that for a material of compressibility κ,

$$|\kappa^{-1}(\partial \ln T_c/\partial P - \partial \ln \sigma_{000}/\partial P)| = |\partial \ln T_c/\partial \ln V - \partial \ln \sigma_{000}/\partial V|$$

is of order unity, although $\partial \ln T_c/\partial \ln V$ and $\partial \ln \sigma_{000}/\partial \ln V$ may be ~ 100.

While a Hartree–Stoner–Wohlfarth theory of the exchange splitting is adequate to explain the ground state of a weak ferromagnet, a more refined theory is in general necessary for the temperature dependence of the magnetization and the pressure dependence of the Curie point. In Stoner theory the magnetic excitations are electron–hole pairs of opposite spin which move independently through the lattice. Moriya showed that correlated motion of the electron–hole pairs leads to different thermodynamic results from those predicted by Stoner theory, e.g. Moriya and Kawabata (1973). The correlated motion is described in terms of spin density fluctuations which are predicted to be particularly important in the case of weak ferromagnetism.

The self-consistent renormalized theory of spin fluctuations of Moriya was subsequently extended by Lonzarich and Taillefer (1985) who showed that a knowledge of four ground state parameters is sufficient to predict the temperature dependence of the magnetization and the value of the Curie point of a weak ferromagnet such as Ni_3Al. However, two of these parameters are not available for $Y(Co_{1-x}Al_x)_2$ because it has not yet proved possible to perform neutron scattering experiments on this system. Takahashi (1986) has stressed the importance of zero point spin fluctuations in weak ferromagnetic materials and derived an expression for the Rhodes–Wohlfarth ratio, section 3, but the theory was not developed to the point where the volume dependence of the magnetization could be evaluated.

Mohn *et al* (1987) and Mohn and Wohlfarth (1987) used the spin fluctuation theory of Lonzarich and Taillefer (1985) and the Landau theory of phase transitions to obtain an equation for the Curie temperature

$$(T_c/T_c^S)^2 + T_c/T_{SF} - 1 = 0$$

where T_c^S is the Curie temperature calculated from Stoner theory and T_{SF} the spin fluctuation temperature which may be calculated from properties of the magnetic ground state. When $T_c^S \gg T_{SF}$, as is expected for a weak ferromagnet, then T_c is determined by spin fluctuations rather than single-particle excitations.

Mohn *et al* (1987) obtain a complicated general expression for the pressure dependence of the Curie point of a band ferromagnet in terms of $t_c = T_c/T_c^S$ which reduces to the simple form

$$\mathrm{d}T_c(P)/\mathrm{d}P = -T_c(0)/P_c \qquad (t_c^2 \ll 1) \tag{1}$$

where $T_c(P_c) = 0$ K.

In contrast both Stoner–Wohlfarth theory and the original form of Moriya's theory predict a non-linear equation for T_c of the form

$$T_c(P) = T_c(0)(1 - P/P_c)^\tau$$

with $\tau = \frac{1}{2}$ (Stoner) or $\frac{3}{4}$ (Moriya), e.g. Wohlfarth (1981).

It will be seen in section 5 that the experimental results for T_c as a function of pressure for the Y–Co–Al alloys are in agreement with the form of equation (1).

2.2. YCo_2

The general features of the density of states of YCo_2 have been known for some ten years (see Yamada (1988) for a useful review), but it will be seen that some refinements have been introduced recently. Cyrot and Lavanga (1979), using the moment method, first showed that the Fermi level of YCo_2 lies on a sharply falling portion of the density of states curve, $D(\varepsilon)$, as a function of energy, figure 1(*a*). Yamada (1988) and Mohn and Schwarz (1990) then showed that there is more structure in $D(\varepsilon)$ near ε_F, figure 1(*b*), with a narrow peak just below the Fermi level. While YCo_2 is only a Pauli paramagnet it was predicted by Cyrot and Lavanga (1979) that a metamagnetic transition could occur in a field estimated at 100 T. Improved calculations reduced the calculated value of the critical field and finally Yamada and Shimizu (1985) estimated it to be 89 T.

The metamagnetic transition in YCo_2 has now been observed at a surprisingly low critical field, 70 T at 10 K, by Goto *et al* (1989). Yamada and Shimizu (1989) refined their earlier calculation to allow for the change of volume of YCo_2 in an external field

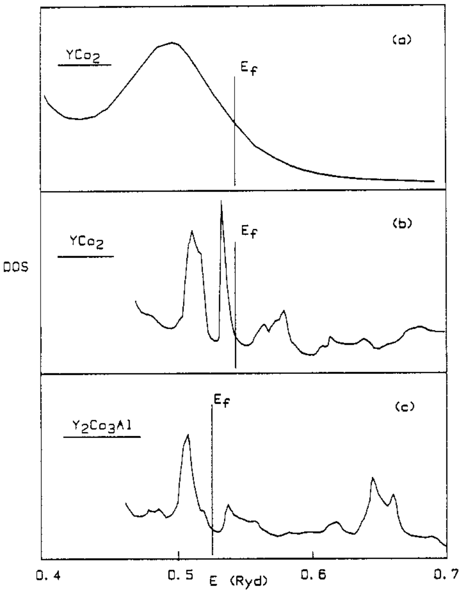

Figure 1. Schematic of the density of states near the Fermi level for (*a*) YCo_2, after Cyrot and Lavanga (1979), (*b*) YCo_2 and (*c*) ordered $Y(Co_{0.75}Al_{0.25})_2$ after Aoki and Yamada (1989) and Mohn and Schwarz (1990).

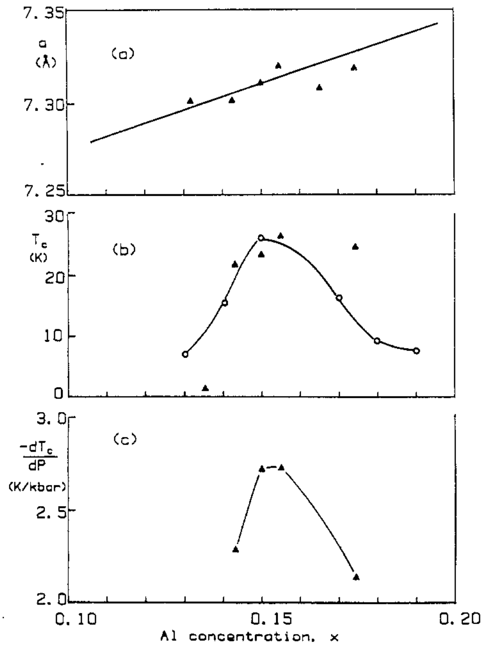

Figure 2. (*a*) The lattice constants, (*b*) the Curie points and (*c*) the pressure dependence of the Curie points of $Y(Co_{1-x}Al_x)_2$ as a function of x. Full lines in (*a*) and (*b*) are from Yoshimura and Nakamura (1985) and the triangles are for the present samples. The line in (*c*) is merely a guide to the eye.

and found that the magnetovolume effect was sufficient to reduce the calculated value of the critical field from 89 T to ~76 T.

2.3. $Y(Co_{1-x}Al_x)_2$

Yoshimura and Nakamura (1985) found that $Y(Co_{1-x}Al_x)_2$ exhibited weak ferromagnetism for $0.12 < x < 0.20$. The maximum spontaneous moment at 0 K was about 0.14 μ_B/Co atom and the maximum Curie temperature ~25 K for $x \sim 0.15$. A comparison of the experimental results for $Y(Co_{1-x}Al_x)_2$ with the spin fluctuation theory of weak itinerant ferromagnetism is given in the next section.

The effect of Al substitution is to increase the lattice constant of YCo_2, figure 2, and to decrease the number of d electrons. In a rigid band model, using figure 1(*a*), both effects would act to increase $D(\varepsilon_F)$ and hence to satisfy the Stoner criteria, $ID(\varepsilon_F) > 1$, discussed in section 2.1. Although no calculations have been performed for random Y–Co–Al alloys, the calculations for the ordered compound $Y(Co_{0.75}Al_{0.25})_2$, Aoki and Yamada (1989), Mohn and Schwarz (1990) show that the reality is more complicated than a rigid band model, figure 1(*c*). (Note that the results shown in figures 1(*b*) and 1(*c*) were calculated using the experimental lattice constants). The hybridization between Co 3d states and Al 3p states is so strong in ordered $Y(Co_{0.75}Al_{0.25})_2$ that one of the peaks in the density of states of YCo_2 is broadened and pushed above ε_F. The value of $D(\varepsilon_F)$ for $Y(Co_{0.75}Al_{0.25})_2$ is actually lower than for YCo_2 so it is calculated to be paramagnetic, in agreement with experiment. In the range $0.12 < x < 0.20$ it must be supposed that ε_F crosses the broad peak observed just above ε_F in figure 1(*c*) and $ID(\varepsilon_F)$ then satisfies the criteria for ferromagnetism. A further discussion of figure 1(*c*) is given in section 5.

3. Previous experiments on $Y(Co_{1-x}Al_x)_2$

Although the first observation of ferromagnetism in $Y(Co_{1-x}Al_x)_2$ was only reported by Yoshimura and Nakamura (1985), the literature is already large and only the main features of the system are listed here. In the original paper Yoshimura and Nakamura showed that, for $0.12 < x < 0.20$, $Y(Co_{1-x}Al_x)_2$ exhibited the following classic features of a weak itinerant ferromagnet:

(i) a large Rhodes–Wohlfarth factor i.e. $p_{eff}/p_0 \gg 1$, where p_{eff} is the moment per Co atom deduced from the high temperature susceptibility and p_0 the spontaneous moment at 0 K, Rhodes and Wohlfarth (1963);

(ii) linear Arrott plots at all compositions, i.e. σ^2_{TH0} versus H/σ_{TH0}, at a given temperature. A linear Arrott plot is usually interpreted as showing a magnetically homogeneous material. Curved lines have frequently been observed in particular samples of e.g. Ni_3Al or MnSi which are, in principle, good weak ferromagnets;

(iii) near T_c the temperature dependence of the spontaneous magnetization and inverse of the paramagnetic susceptibility follow a $|T_c^{4/3} - T^{4/3}|$ law as predicted by spin fluctuation theory.

High field magnetization measurements on $Y(Co_{1-x}Al_x)_2$ (Sakakibara *et al* 1987, Wada *et al* 1988) showed metamagnetic transitions for $x < 0.12$ and a small spontaneous moment which increases rapidly in an applied field for $x > 0.13$. This strongly field dependent magnetization will be seen in section 5 be important for the interpretation of our forced magnetostriction data.

Microscopic measurements have also confirmed the weak itinerant ferromagnetism of $Y(Co_{1-x}Al_x)_2$. NMR experiments (Yoshimura *et al* 1987, 1988) on both ^{27}Al and ^{59}Co showed that all Co atoms were equivalent, i.e. there was no evidence for a Co atom only carrying a magnetic moment if surrounded by more than a certain number of Co neighbours. The temperature and magnetic field dependence of the spin–lattice relaxation rate was found to be in agreement with the spin fluctuation theory of Takahashi (1986). Similarly, muon spin rotation experiments, which probe the interstitial magnetization, carried out in transverse geometry in a field of 0.02 T, i.e. much smaller than the field required for NMR, showed critical damping of the muon relaxation rate at a temperature in good agreement with the T_c found from AC susceptibility measurements (Graham *et al* 1989).

4. Experimental details

Alloys with compositions $x = 0.135, 0.143, 0.15, 0.155, 0.165$ and 0.175 were prepared from 99.99% pure Y and Co and 99.999% pure Al by argon arc melting; the samples were turned and re-melted to ensure homogeneity. The arc melted buttons were then

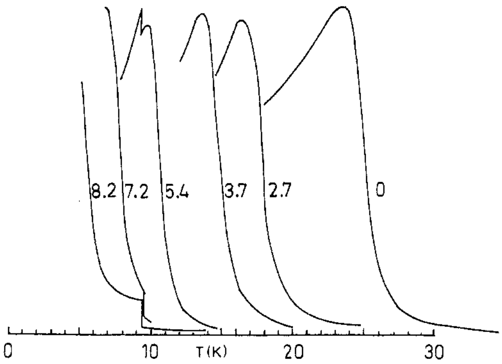

Figure 3. The AC susceptibility (in arbitrary units) of $Y(Co_{0.845}Al_{0.155})_2$ as a function of temperature at pressures between atmospheric and 8.2 kbar. The step near 9 K is due to the superconducting transition in Nb (phase reversed by the arrangement of the mutual inductance.)

Table 1. The Curie point, pressure dependence of the Curie point and critical pressure, i.e. the pressure at which T_c extrapolates to zero, equation (1), for $Y(Co_{1-x}Al_x)_2$.

x	$T_c(0)$ (K)	$dT_c(P)/dP$ (K kbar)	P_c (kbar)
0.143	21.4	−2.4	8.9
0.150	23.1	−2.6	8.9
0.150†	21.4	−2.1	10.0
0.155	26.0	−3.0	8.7
0.175	24.2	−2.2	11.0

† Sample provided by Professor Yoshimura.

wrapped in Ta foil and sealed under argon in silica tubes and annealed at 950 °C for 14 days. X-ray diffraction patterns recorded from as-crushed powders showed the alloys to be single phase except for the occurrence of a few very weak extra lines in the case of the two highest Al content samples. The lattice parameters were in good agreement with previous values (Yoshimura and Nakamura 1985), as shown in figure 2.

The Curie points of the alloys were measured, using a mutual inductance technique, on rod shaped samples which were spark cut from the original ingots. While the AC susceptibility is not a rigorous method for determining T_c it was the shift of T_c with pressure which was of most interest in the present experiments and since there was no evidence of the transition broadening under pressure, figure 3, reliable values of dT_c/dP were obtained. The Curie point was defined as the temperature at which the AC susceptibility showed a maximum slope. Values of T_c at atmospheric pressure as a function of composition are shown in figure 2 and table 1. The agreement with the results of Yoshimura and Nakamura (1985) is good near $x = 0.155$ but our values of T_c tend to be higher than theirs for $x > 0.15$ and lower for $x < 0.15$. There was no evidence for a magnetic transition down to 4 K in our $x = 0.135$ sample.

The mutual inductance containing the sample was placed in a liquid filled Be–Cu pressure cell. Pressure up to 10 kbar was applied at room temperature and locked into the cell. A gas flow cryostat was used to cool the cell to a minimum temperature of 6 K. The pressure inside the cell, up to 8 kbar at low temperature, was measured using a calibrated semiconductor transducer. The temperature of the cell was measured by a carbon glass thermometer placed in a recess at the top of the pressure cell. The cell was slowly cycled between 6 K and a few degrees above T_c until consistent values were obtained for T_c on warming and cooling. Values of T_c measured in this manner with the sample in the pressure cell at atmospheric pressure were in good agreement with measurements made with the thermometer and sample side by side in a copper block. As a further check on the temperature inside the cell a piece of Nb was inserted in the mutual inductance since the superconducting transition of Nb (9.2 K) is nearly independent of pressure.

A typical series of measurements of the AC susceptibility as a function of temperature at various pressures is shown in figure 3. The peak in the AC susceptibility below T_c, and the fact that the transition region is only about 2 K wide, suggest that the material is of high quality. It will be apparent from the strong pressure dependence of T_c shown in figure 3 that weakly ferromagnetic materials must be extremely sensitive to non-uniform strain, a good sample of $ZrZn_2$ for example was found to have a transition width of 4 K and showed no peak in the AC susceptibility below T_c, so this $Y(Co_{0.845}Al_{0.145})_2$ sample must be both magnetically homogeneous and strain free. The sample with $x = 0.165$ appeared to be of less uniform composition and had a transition width of ~4 K. A $Y(Co_{0.85}Al_{0.15})_2$ sample provided by Professor Yoshimura was found to have a transition width of 5 K.

The forced magnetostriction was measured on discs of diameter 5 mm and thickness 1 mm which were spark cut from the same ingots as the rods used for the T_c measurements. Fields of up to 12 T at 4.2 K or 5 T at 1.6 K were applied in the plane of the disc. The change of length of the sample perpendicular and parallel to the direction of the field was measured using a capacitance technique with a reproducibility, on cycling the magnetic field, of 3 Å; see Armitage *et al* (1985, 1989) for details. The volume change of the discs was calculated from the equation

$$\Delta V/V = 2\,\Delta l_{\perp}/l_0 + \Delta l_{\parallel}/l_0. \tag{2}$$

There was found to be no significant change in the forced volume magnetostriction between 1.6 K and 4.2 K.

5. Discussion

The change in the Curie point with pressure of each sample of $Y(Co_{1-x}Al_x)_2$ is shown in figure 4. The Curie point is a linear function of pressure, as predicted by equation (1), although there is a small possible deviation from linearity for $x = 0.150$ and 0.155 at the highest pressures, where $T_c(P)$ is close to 6 K, the lowest temperature available in the flow cryostat. Values of $\mathrm{d}T_c(P)/\mathrm{d}P$ as a function of composition are shown in figure 1 and of the critical pressure P_c, as defined in equation (1), in table 1. The estimated error in each value of P_c is ±1 kbar so there is no definite change of P_c in $Y(Co_{1-x}Al_x)_2$ for $0.143 < x < 0.175$.

The average value of P_c for all the samples in table 1 is 9.5 ± 1.0 kbar so from equation (1) and the value $9.4 \times 10^{-4}\ (\text{kbar})^{-1}$ for the isothermal compressibility used by Yamada and Shimizu (1989) the initial decrease in T_c may be written as

$$\partial \ln T_c/\partial \ln V = 112 \pm 11 \tag{3}$$

which is comparable to that found for the typical weak ferromagnet $ZrZn_2$ (Fawcett and Meincke 1971). A further discussion of equation (3) will be given after a consideration

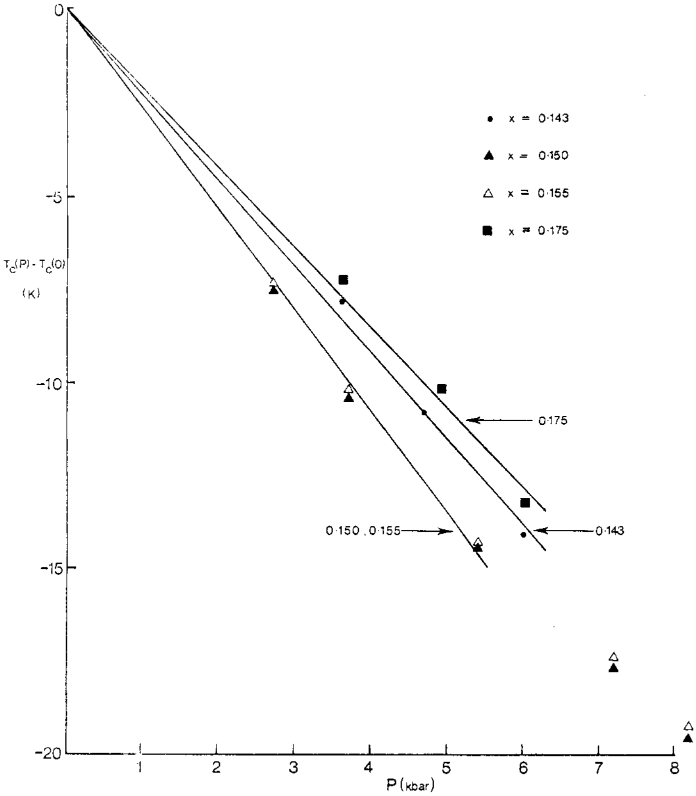

Figure 4. The change in the Curie point, from that at atmospheric pressure of $Y(Co_{1-x}Al_x)_2$ with pressure.

of the forced magnetostriction data from which it will be shown that $\partial \ln T_c / \partial \ln V \sim \partial \ln \sigma_{000} / \partial \ln V$ for the Y–Co–Al system.

The forced magnetostriction at 4.2 K of the $Y(Co_{1-x}Al_x)_2$ samples with $0.135 < x < 0.175$ was found to be a non-linear function of the magnetic field, figure 5. However, apart from the 0.135 sample, which appeared to be paramagnetic, an adequate fit to the curves was given by the simple expression

$$(l_H - l_0)/l_0 = aH + bH^2 \tag{4}$$

where a and b were constants for a given sample and orientation of the field. It has often been assumed in magnetostriction studies of weakly ferromagnetic materials that the change of length parallel to a given field was equal to that perpendicular to it. The present measurements are sufficiently accurate to show, table 2, that for $Y(Co_{1-x}Al_x)_2$, $a_\perp \sim 1.2 a_\parallel$.

The non-linear behaviour of l_H with H expressed by equation (4) is not observed in YFe_2 (Armitage *et al* 1985, 1989) because the magnetization of YFe_2 saturates above 2 T. The magnetization of $Y(Co_{1-x}Al_x)_2$, however, is a strong function of the magnetic field; e.g. for $x = 0.15$ the magnetization at 10 T is 2.5 times the spontaneous magnetization (Sakakibara *et al* 1987). Spin fluctuation theory predicts that

$$(V_H - V_0)/V_0 = \Delta V/V_0 = C\kappa(\sigma^2_{THP} - \sigma^2_{T0P}) \tag{5}$$

where C is the magneto-volume coupling constant and κ the isothermal compressibility.

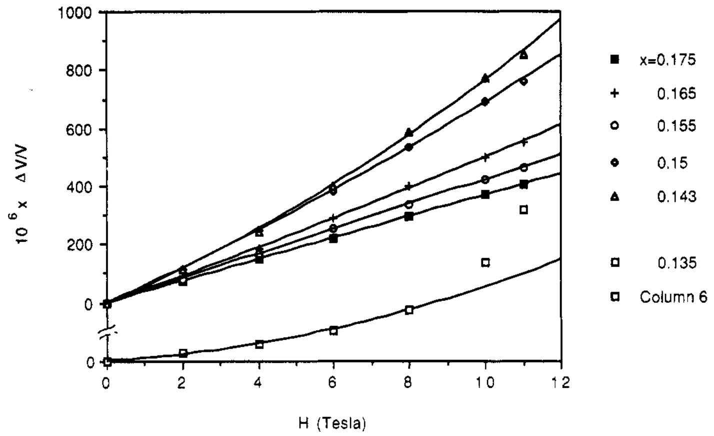

Figure 5. The fractional change in volume of $Y(Co_{1-x}Al_x)_2$ with magnetic field at 4.2 K. The lines are calculated using equations (2) and (4) and the data of table 2.

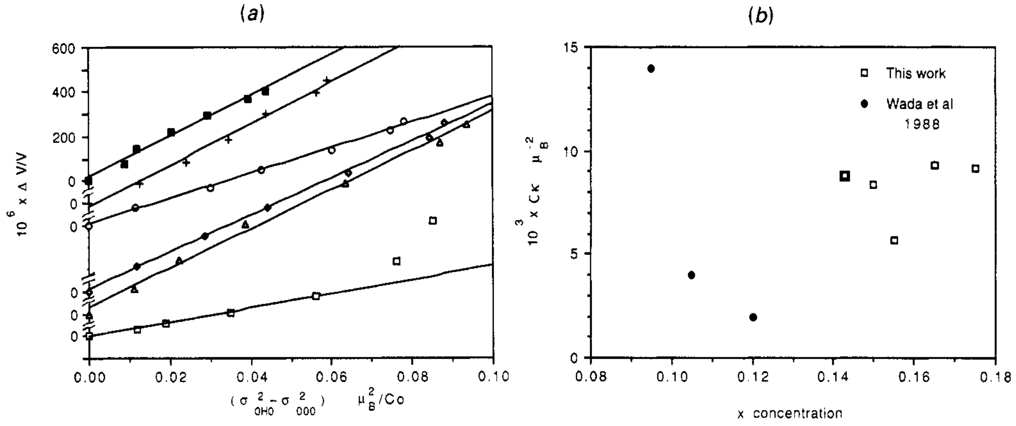

Figure 6. (*a*) The change of volume of $Y(Co_{1-x}Al_x)_2$ as a function of $(\sigma^2_{0H0} - \sigma^2_{000})$; see equation (5). (*b*) The product of the magnetovolume coupling constant and the isothermal compressibility as a function of composition for $Y(Co_{1-x}Al_x)_2$ (□, 0.135; △, 0.143; ◇, 0.150; ○, 0.155; +, 0.165; ■, 0.175).

Equation (5) was found to be satisfied by paramagnetic $Y(Co_{1-x}Al_x)_2$ with $0.095 < x < 0.12$ (Wada *et al* 1988). We do not have magnetization data for the samples on which the magnetostriction experiments were performed, and there are some differences between the high field magnetization data of Wada *et al* (1988) and Sakakibara *et al* (1987), as would be expected for weak ferromagnets, but as shown in figure 6(*a*), equation (5) produced reasonable straight lines for weakly ferromagnetic $Y(Co_{1-x}Al_x)_{2T}$. The coupling constant $C\kappa$, where the data of Sakakibara *et al* (1987) have been used for both paramagnetic and ferromagnetic $Y(Co_{1-x}Al_x)_2$ as a function of composition is shown in figure 6(*b*). The value of $C\kappa$ decreases from a rather large value for $x = 0.12$ and then appears to be almost constant in the ferromagnetic phase with a value similar to that of $GdCo_2$ (Muraoka *et al* 1984).

Table 2. The constants a and b, as defined in equation (4), for the forced linear magnetostriction of $Y(Co_{1-x}Al_x)_2$ with the field parallel or perpendicular to the direction of measurement. The critical pressure, as defined in equation (7), is also given, where the magnetization data of Yoshimura and Nakamura (1985) has been used. The product of the magnetovolume coupling constant and the isothermal compressibility has been calculated from equation (5) using the high field magnetization data of Sakakibara *et al* (1987).

	10^6a		10^8b			
x	$a_\parallel$	$a_\perp$	$b_\parallel$	$b_\perp$	P_c (kbar)	$10^3C\kappa$ $((\mu_B/Co)^{-2})$
0.135	5.3	3.1	224	58	—	—
0.143	14.2	16.9	92	109	8.2	8.82
0.150	15.3	18.3	58	65	8.2	8.34
0.155	12.4	14.4	7	6	10.5	5.71
0.165	12.5	15.7	31	22	7.9	9.35
0.175	10.8	13.2	1	−1	6.7	9.19

The pressure dependence of the magnetization per unit mass can be found from the forced magnetostriction by using the thermodynamic relation,

$$(\partial\sigma/\partial P)_{TH} = -(1/\rho)(\partial \ln V/\partial H)_{TP} \tag{6}$$

where ρ is the density. In a weak ferromagnet, where the magnetization is a strong function of H, it will be the term linear in H in equation (4) which relates to the spontaneous magnetization, i.e.

$$\partial\sigma_{000}/\partial P = -(a_\parallel + 2a_\perp)/\rho.$$

In order to estimate the pressure at which the spontaneous magnetization at 0 K goes to zero we write, as a linear extrapolation from low pressure,

$$P_c = \sigma_{000}\, \partial P/\partial\sigma_{000} = -\rho\sigma_{000}/(a_\parallel + 2a_\perp). \tag{7}$$

(Since the values of $a_\parallel$ and $a_\perp$ were found not to change between 4.2 K and 1.6 K they do not need to be corrected to 0 K.)

Values of σ_{000} were taken from Yoshimura and Nakamura (1985) and the derived values of P_c for the $Y(Co_{1-x}Al_x)_2$ are shown in table 2. It will be seen that, not surprizingly, there is a greater range of values of P_c in table 2 than in table 1, but the values of P_c probably do not differ significantly from the average value of 8.7 ± 1.2 kbar obtained for $0.143 \leq x \leq 0.165$. The low value of P_c (6.7 kbar) deduced for $x = 0.175$ probably arises because the magnetization of our sample is greater than that of Yoshimura and Nakamura (1985). The ratio of the Curie points for the two samples is ~1.5, and since in weak ferromagnets σ_{000} and T_c tend to scale together, as found e.g. for Ni_3Al (Sasakura *et al* 1984), this would increase the value of P_c for the present $x = 0.175$ sample to ~10 kbar. We cannot, however, exclude a compositional dependence of the critical pressure and direct measurements of the magnetization as a function of pressure, rather than the use of forced magnetostriction data, would be very desirable.

It will be seen from table 1 and table 2 that there is no significant difference between the pressure dependence of the Curie point and the spontaneous magnetization at 0 K for $Y(Co_{1-x}Al_x)_2$ and an overall average leads to the result for $0.143 < x < 0.175$,

$$\partial \ln T_c/\partial \ln V = \partial \ln \sigma_{000}/\partial \ln V = 120 \pm 17 \tag{8}$$

which is negligibly different from equation (3).

The equality of $\partial \ln T_c/\partial \ln V$ and $\partial \ln \sigma_{000}/\partial \ln V$ for $Y(Co_{1-x}Al_x)_2$, to the accuracy of equation (7), is in agreement with the theory of weak ferromagnetism; e.g. Wohlfarth (1981) suggests that the difference between $\partial \ln T_c/\partial \ln V$ and $\partial \ln \sigma_0/\partial \ln V$ is of order unity. It has recently, however, been confirmed for Sc_3In, often quoted as a typical weak ferromagnet, that

$$\partial \ln T_c/\partial \ln V \sim 3.3\, \partial \ln \sigma_{000}/\partial \ln V \sim -45$$

so $\partial \ln T_c/\partial \ln V$ is not equal to $\partial \ln \sigma_{000}/\partial \ln V$ and the derivatives are actually negative for pressures up to at least 31 kbar. (Earlier suggestions on theoretical grounds (Wagner and Wohlfarth 1981) that the negative value for $\partial \ln T_c/\partial \ln V$ for Sc_3In was due to heterogeneity can no longer be sustained.)

It was seen in section 2 that computer calculations suggest that the addition of Al to YCo_2 broadens a narrow peak in the density of states near the Fermi level of YCo_2 and in the artificial case of ordered $Y(Co_{0.75}Al_{0.25})_2$ moves that peak above the Fermi level, figure 1. The density of states curve for ordered $Y(Co_{0.75}Al_{0.25})_2$ is rather misleading for random $Y(Co_{1-x}Al_x)_2$ because the randomness would also broaden a second peak shown below the Fermi level in figure 1(*c*).

While it is clear that a rigid band model is not correct between YCo_2 and ferromagnetic $Y(Co_{1-x}Al_x)_2$ it is more reasonable to think that such a model would be appropriate for the consideration of the volume dependence of the magnetization and Curie point at a given value of x. The fact that the critical pressure for $Y(Co_{1-x}Al_x)_2$ in the range $0.143 < x < 0.175$ is constant (to ~ 10%) can then be understood if it is assumed that the width of the peak in the density of states near the Fermi level increases with increasing x but its maximum value decreases and moves to higher energy. The variation of the lattice constant between YCo_2 and $Y(Co_{1-x}Al_x)_2$ shown in figure 2(*a*) may be represented as a negative chemical pressure,

$$-\Delta P\,(\mathrm{kbar}) = -266x.$$

Since the critical pressure to destroy ferromagnetism is $P_c \sim 9$ kbar it is clear that ferromagnetism, observed only for $0.12 < x < 0.20$ at atmospheric pressure, would be impossible in the $Y(Co_{1-x}Al_x)_2$ system at the volume of YCo_2. The change of chemical pressure between the optimum value of x for ferromagnetism, ~0.155, and $x = 0.12$ is however roughly equal to P_c.

6. Conclusion

Earlier measurements of both bulk and microscopic (NMR, μSR) properties of $Y(Co_{1-x}Al_x)_2$ suggested that weak itinerant ferromagnetism occurred for $0.12 < x < 0.20$. It has now been shown that, for $0.143 < x < 0.175$, $Y(Co_{1-x}Al_x)_2$ also satisfies the theoretical criterion

$$\partial \ln T_c/\partial P = \partial \ln \sigma_{000}/\partial P < 0.$$

The values of $\partial \ln T_c/\partial \ln V$ and $\partial \ln \sigma_{000}/\partial \ln V$ are large and positive (120 ± 17), as found for the well established weak ferromagnet $ZrZn_2$ but in contrast to the findings for Sc_3In. The critical pressure to reduce the Curie point to 0 K or the spontaneous magnetization at 0 K to zero was found to be 9 ± 1 kbar, independent of the value of x, table 1 and 2.

Although $Y(Co_{1-x}Al_x)_2$ is a random alloy the sharpness of the magnetic transition at the Curie point and the lack of sensitivity of the magnetic properties to small fluctuations in composition near the optimum value of $x = 0.15$ is superior to that of $ZrZn_2$, Ni_3Al or Sc_3In. In Ni_3Al for example (Sasakura *et al* 1984) increasing the nickel content by only one per cent changes the Curie point from 23 K to 58 K and the spontaneous magnetization by a similar factor and in Sc_3In (Grew *et al* 1989) T_c may vary by 50% with composition. In contrast samples of $Y(Co_{0.85}Al_{0.15})_2$ prepared in different laboratories have been found to have T_c different by only some 8%, table 1, and values of P_c of 9 ± 1 and 10 ± 1 kbar. The $Y(Co_{1-x}Al_x)_2$ system is therefore a valuable addition to the small number of materials which appear to satisfy all the theoretical criteria for weak itinerant ferromagnetism.

Acknowledgments

We are indebted to Dr P Mohn for helpful discussions of the theory of weak ferromagnetism and for details of his calculation with Professor K Schwarz of the density of states of $Y(Co_{0.75}Al_{0.25})_2$. Professor K Yoshimura kindly provided samples of $Y(Co_{1-x}Al_x)_2$.

References

Aoki M and Yamada H 1989 *J. Magn. Magn. Mater.* **78** 377–83
Armitage J G M, Dumelow T, Mitchell R H, Riedi P C, Abell J S, Mohn P and Schwarz K 1986 *J. Phys. F: Met. Phys.* **16** L141–4
Armitage J G M, Dumelow T, Riedi P C and Abell J S 1989 *J. Phys.: Condens. Matter* **1** 3987–94
Cyrot M and Lavanga M 1979 *J. Physique* **40** 763–71
Fawcett E and Meincke P P M 1971 *J. Physique* **32** C1 629–31
Goto T, Fukamichi K, Sakakibara T and Komatsu H 1989 *Solid State Commun.* **72** 945–7
Graham R G, Armitage J G M, Riedi P C, Cox S F J and Scott C A 1989 *Muon Studies in Solid State Physics (IOP Short Meetings Series No 22)* ed S F J Cox (Bristol: Institute of Physics) pp 41–4
Grew J, Schilling J S, Ikeda K and Gschneider Jr K A 1989 *Phys. Rev.* B **40** 9017–24
Lonzarich G G and Taillefer L 1985 *J. Phys. C: Solid State Phys.* **18** 4339–71
Mohn P and Schwarz K 1990 Private communication
Mohn P, Wagner W and Wohlfarth E P 1987 *J. Phys. F: Met. Phys.* **17** L13–8
Mohn P and Wohlfarth E P 1987 *J. Phys. F: Met. Phys.* **17** 2421–30
Moriya T and Kawabata A 1973 *J. Phys. Soc. Japan* **35** 669–76
Rhodes P and Wohlfarth E P 1963 *Proc. R. Soc.* **273** 247–58
Riedi P C, Armitage J G M, Graham R G and Abell J S 1988 *J. Physique* **48** C8 269–70
Sakakibara T, Goto T, Yoshimura K, Shiga M, Nakamura Y and Fukamichi K 1987 *J. Magn. Magn. Mater.* **70** 126–8
Sasakura H, Susuki K and Masuda Y 1984 *J. Phys. Soc. Japan* **53** 754–9
Takahashi Y 1986 *J. Phys. Soc. Japan* **55** 3553
Wada H, Yoshimura K, Kido G, Shiga M, Mekata M and Nakamura Y 1988 *Solid State Commun.* **65** 23–6
Wohlfarth E P 1981 *Physics of Solids under High Pressure* ed J S Schilling and R N Shelton (Amsterdam: North-Holland) pp 175–80
Wagner D and Wohlfarth E P 1981 *J. Phys. F: Met. Phys.* **11** 2417–28
Yamada H 1988 *Physica* B **149** 390–402
Yamada H and Shimizu M 1989 *Physica* B **161** 179–82
Yoshimura K and Nakamura Y 1985 *Solid State Commun.* **56** 767–71
Yoshimura K, Takigawa M, Takahashi Y, Yasuoka H and Nakamura Y 1987 *J. Phys. Soc. Japan* **56** 1138–55
Yoshimura K, Mekata M, Takigawa M, Takahashi Y and Yasuoka H 1988 *Phys. Rev.* B **37** 3593–602