Electrical resistivity and the band structure of $Fe_3Si_{1-x}Al_x$ alloys

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(Received 9 July 1981)

The electrical resistivity of a series of $Fe_3Si_{1-x}Al_x$ alloys with $0 \leq x \leq 1$ has been measured from 1.8 to 300 K. $(d\rho/dt)|_{300\,K}$ was found to vary less than 20% over the composition range, while the residual resistivity ρ_0 increased sixfold in going from Fe_3Si to Fe_3Al. We argue that the constancy of $(d\rho/dt)|_{300\,K}$ implies that the density of states at the Fermi level is due largely to Fe atoms, thus confirming recent band-structure calculations. We also show that the increase in ρ_0 can be accounted for in terms of the Fe-Al site disorder in Fe_3Al.

INTRODUCTION

This paper is an extension of our work on the magnetic properties and hyperfine field distributions in $Fe_3Si_{1-x}Al_x$ alloys[1] and our work on the electrical resistivity of $Fe_{3+x}Si_{1-x}$.[2] Here we present measurements of the electrical resistivity between 1.8 and 300 K on a series of $Fe_3Si_{1-x}Al_x$ alloys with composition in the range $0 \leq x \leq 1$.

Both Fe_3Si and Fe_3Al have the DO_3 crystal structure shown in Fig. 1. This structure can be viewed as a Heusler-type alloy which has the $L2_1$ structure with the inequivalent (A,C) and (B) sites both occupied by Fe and the (D) site occupied randomly by Si or Al. All the materials are ferromagnetic and a summary of their magnetic and crystallographic properties is given in Table I and Ref. 1.

Sample preparation and measurements

The samples were prepared by arc melting under titanium gettered argon. Those ingots with $x < 0.15$ were then annealed at 800 °C for 4 h and furnace cooled to 600 °C then annealed for 1 h and funace cooled to room temperature. Resistance samples $2 \times 2 \times 16$ mm^3 were cut from the ingot with a low speed diamond saw. These samples were then given a stress relieving anneal at 600 °C for one hour and furnace cooled to room temperature. The ingots with $x > 0.15$ were annealed at 950 °C for 3 days and quenched in ice water. Resistance samples were cut from the ingot and annealed at 500 °C for 4 h and slow cooled to room temperature. These annealing prescriptions were previously determined to produce highly ordered specimens.[1]

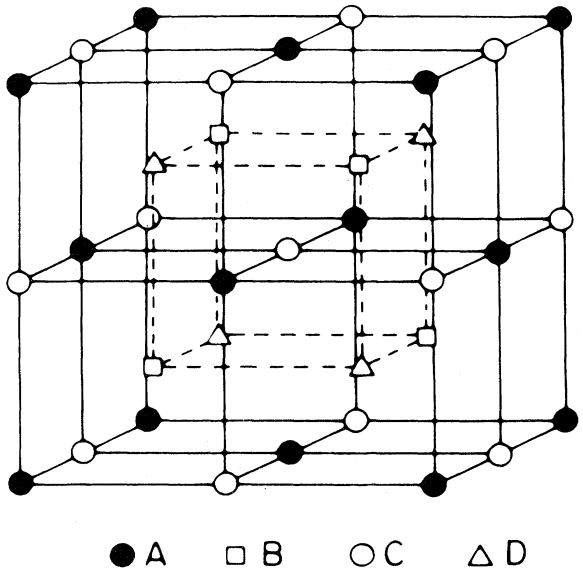

FIG. 1. Unit cell of the $L2_1$ structure. This reduces to the DO_3 structure when the A and C sites are identical which is the case for Fe_3Si and Fe_3Al. In the present alloys A, B, and C are occupied by Fe and D by Si and/or Al.

TABLE I. Properties of Fe_3Si and Fe_3Al.

	Fe_3Si	Ref.	Fe_3Al	Ref.
Crystal structure	DO_3	1	DO_3	1
Lattice constant	5.553 Å	1	5.793 Å	1
Magnetic structure	Ferromagnetic	1	Ferromagnetic	1
Curie temperature	840 K	1	713 K	1
Moments	$Fe(B)=2.2\mu_B$	1	$Fe(B)=2.2\mu_B$	1
	$Fe(A,C)=1.35\mu_B$	1	$Fe(A,C)=1.45\mu_B$	1
	$Si(D)=0\mu_B$	1	$Al(D)=0\mu_B$	1
Internal Fields	$Fe(B)=338$ kOe	1	$Fe(B)=330$ kOe	1
	$Fe(A,C)=218$ kOe	1	$Fe(A,C)=234$ kOe	1
	$Si(D)=37$ kOe	1	$Al(D)=26$ kOe	1
Average atomic mass M $=\frac{1}{4}$(mass per formula unit)	48.90 g		48.63 g	

The resistance was measured using an ac difference technique.[3] The resistivity of the samples was measured at 293 K in a room-temperature jig which ensured accurate contact spacing. The results of the measurements are shown in Fig. 2. The most striking feature of the results is the great similarity of the thermal part of the resistivity across the whole concentration range. This is demonstrated in Fig. 3(a) where $(d\rho/dT)\,|_{300\,K}$ is plotted as a function of x along with some preliminary results for the $Fe_{3-x}Co_xSi$ alloy system. The residual resistivity, on the other hand, increases more than sixfold on going from Fe_3Si to Fe_3Al as shown in Fig. 3(b).

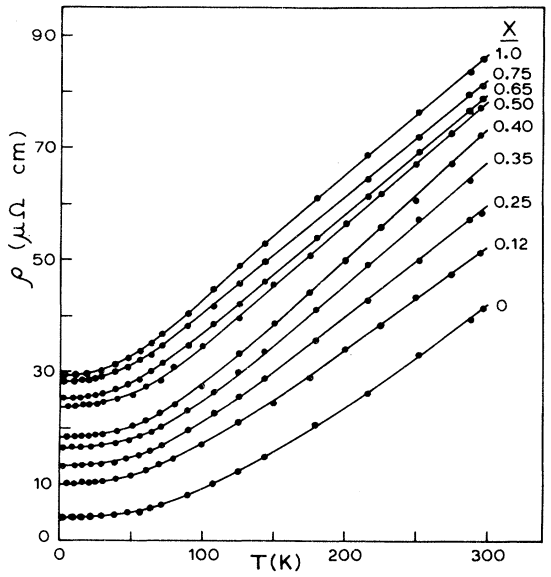

FIG. 2. ρ as a function of temperature T and composition x for $Fe_3Si_{1-x}Al_x$ alloys.

DISCUSSION

The electrical resistivity of a metal is given by Ziman[4] as

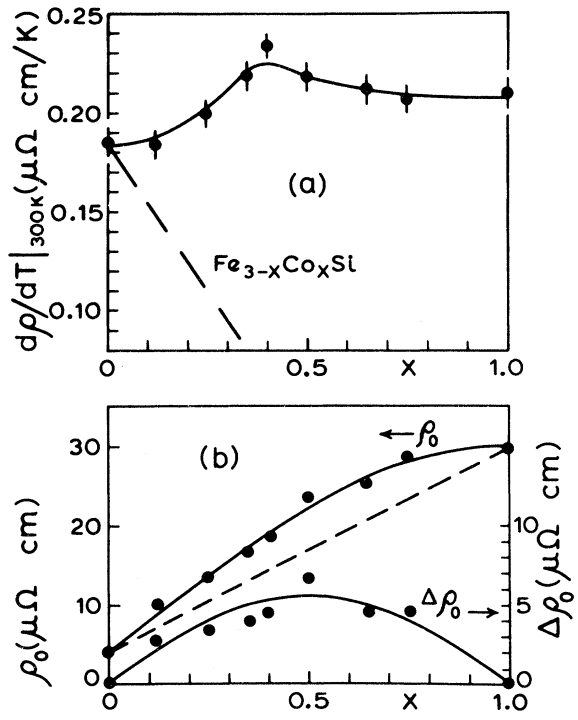

FIG. 3. (a) $(d\rho/dT)\,|_{300\,K}$ as a function of x for $Fe_3Si_{1-x}Al_x$. The dashed line shows some preliminary data for $Fe_{3-x}Co_xSi$. (b) ρ_0 as a function of x for $Fe_3Si_{1-x}Al_x$. The dashed line is the calculated change in ρ_0 due to Fe-Al site disorder. $\Delta\rho_0$ is the difference between ρ_0 and the dashed line. The solid line is given by $\Delta\rho_0=4x(1-x)$ with $\Delta\rho_0(\text{max})=5.5\ \mu\Omega\,\text{cm}$.

$$\rho=\frac{3}{(eV_F)^2}\frac{1}{N(E_F)}\left[\frac{1}{\tau_0}+\frac{1}{\tau_i(T)}\right], \quad (1)$$

where $N(E_F)$ is the density of states at the Fermi energy, V_F is the Fermi velocity, and τ_0 and $\tau_i(T)$ are the relaxation times due to impurity and phonon scattering, respectively. In the spirit of Matthiessen's rule the two scattering mechanisms are assumed to be independent and τ_0 is assumed to be independent of temperature.[5] The slope of the resistivity curve at high temperatures $(d\rho/dT)\,|_{300\,\mathrm{K}}$, is given by

$$\left.\frac{d\rho}{dT}\right|_{300\,\mathrm{K}}=\frac{3}{(eV_F)^2N(E_F)}\frac{d}{dT}\left[\frac{1}{\tau_i(T)}\right]. \quad (2)$$

The phonon relaxation time at high temperatures from Ref. 6 is

$$\frac{1}{\tau_i(T)}\propto\frac{T}{M\Theta_D^2}$$

and thus

$$\left.\frac{d\rho}{dT}\right|_{300\,\mathrm{K}}\propto[(eV_F)^2N(E_F)M\Theta_D^2]^{-1}, \quad (3)$$

where M and Θ_D are the average ionic mass and the Debye temperature, respectively.

Equation (3) in conjunction with the remarkable constancy of $(d\rho/dT)\,|_{300\,\mathrm{K}}$ across the whole alloying range from Fe_3Si to Fe_3Al along with the virtual constancy of M (see Table I) implies that the product of $V_F^2N(E_F)$ and Θ_D is unchanged on alloying. This in turn implies either that Θ_D and $V_F^2N(E_F)$ are each constant or that they vary in a reciprocal fashion. The latter possibility seems very unlikely and we assume that Θ_D and $V_F^2N(E_F)$ are each constant.[7] The constancy of $V_F^2N(E_F)$ suggests that the electrons involved in the conduction process come from the same source, that is, the one species common to both materials, namely, the Fe atoms. Band-structure calculations for Fe_3Si (Ref. 8) and Fe_3Al (Ref. 9) indeed show that the density of states at the Fermi level in both cases is dominated by the d electrons from the iron atoms. This idea is further reinforced when a comparison is made between the change in $(d\rho/dT)\,|_{300\,\mathrm{K}}$ for $Fe_3Si_{1-x}Al_x$ and $Fe_{3-x}Co_xSi$ with x. The band-structure calculations show that the density of states at the Fermi level is dominated by contributions from the Fe(AC) sites. Co is known to systematically substitute for Fe on the (AC) site and we would therefore expect a large change in $(d\rho/dT)\,|_{300\,\mathrm{K}}$ with Co substitution. The preliminary results shown in Fig. 3(a) strikingly show this to be the case.

The residual resistivity, which from Eq. (1) is given by

$$\rho_0=\frac{3}{(eV_F)^2}\frac{1}{N(E_F)}\frac{1}{\tau_0} \quad (4)$$

is more difficult to explain if $V_F^2N(E_F)$ must remain constant. In this case τ_0 for Fe_3Si must be about six times its value for Fe_3Al.

In our previous work on $Fe_{3+x}Si_{1-x}$ alloys the residual resistivity increased quickly both as Fe entered the Si sites and also as Si entered the Fe sites as is shown in Fig. 4. It is known that there is about an 8% Al-Fe site disorder in Fe_3Al.[10] If we assume that the resistivity due to this disorder corresponds to the sum of the effects observed in the residual resistivity of $Fe_{3+x}Si_{1-x}$ for $|x|=0.04$ then we predict a value of ρ_0 for Fe_3Al which is given by

$$\rho_0Fe_3Al=\rho_0(Fe_{3.04}Si_{0.96})\rho_0(Fe_{2.96}Si_{1.04})$$
$$=28.5\,\mu\Omega\,\mathrm{cm}. \quad (5)$$

This is remarkably close to our measured value of 29.5 $\mu\Omega$ cm for Fe_3Al.

If we further assume that the Al,Fe site disorder is proportional to the amount of Fe_3Al present then ρ_0 should increase linearly with x across the alloy system as indicated by the dashed line in Fig. 3(b). The difference $\Delta\rho_0$ between the dashed line and the measured values of ρ_0 is plotted against x in Fig. 3(b). This difference shows the parabolic form, with a maximum at $x=0.5$, which would be expected for Si-Al disorder scattering. The maximum value of $\Delta\rho_0=5.5$ $\mu\Omega$ cm is typical of the

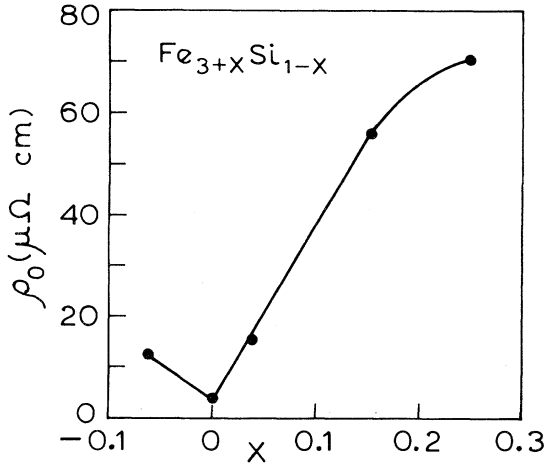

FIG. 4. ρ_0 as a function of x for $Fe_{3-x}Si_x$.

change of resistivity found for this type of scattering.[11]

CONCLUSIONS

The electrical resistivity of a series of $Fe_3Si_{1-x}Al_x$ alloys have been measured for compositions ranging from $x=0$ to $x=1$ over the temperature range 2 to 300 K. The constancy of $(d\rho/dT)\,|_{300\,K}$ as a function of alloy composition in this sytem, where Al substitutes for Si in the D sites, along with the large change observed in $(d\rho/dT)\,|_{300\,K}$ for the $Fe_{3-x}Co_xS_i$ system, where the Co substitutes in the Fe(AC) sites, strongly suggests that electrons from the iron atoms dominate the conduction process. This confirms bandstructure calculations for Fe_3Si (Ref. 8) and Fe_3Al (Ref. 9) which show that the density of states at the Fermi level is almost entirely due to contributions from the Fe(AC) and Fe(B) sites.

The observed sixfold increase in the residual resistivity of Fe_3Al over Fe_3Si is shown to be due to the 8% Fe-Al site disorder in Fe_3Al (Ref. 10). When this effect is accounted for the resistivity due to Si-Al disorder on the D sites is shown to be parabolic in form with a maximum at $x=0.5$. The maximum value of 5.5 $\mu\Omega$ cm for the resistivity due to Si-Al disorder scattering is in reasonable agreement with the values found in other alloy systems.[11]

ACKNOWLEDGMENTS

This work was supported in part by the University of Connecticut Research Foundation and the National Science and Engineering Research Council of Canada. The authors would like to thank P. J. Saunders, L. Masur, M. Discenza, and P. Henry for help with sample preparation and measurements.

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