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Temperature Dependence of Magnetoresistance in Ni3Al System

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Temperature dependence of magnetoresistance in the Ni_3Al system

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The transverse magnetoresistance of stoichiometric and off-stoichiometric polycrystalline samples of the Ni_3Al system has been measured over the temperature range $1.2 < T < 90$ K in magnetic fields up to 23 kOe. Negative magnetoresistance was observed in all cases. In samples with Ni concentrations of 75 at.% or greater the magnitude of the magnetoresistance reached a maximum at $T = T_C$. The data are in qualitative agreement with the theoretical predictions of Chang *et al.* based on a model of the magnetoresistance of an itinerant ferromagnet, but the observed magnetoresistance is considerably larger than predicted.

I. INTRODUCTION

Ni_3Al is a weakly ferromagnetic intermetallic compound. The ferromagnetic transition is accompanied by a large, abrupt change in ac susceptibility and a small but sharp change in specific heat.[1] There is considerable disagreement over the nature of the magnetic ordering in this system. De Boer *et al.*[2] have interpreted their magnetization data on the basis of the Stoner-Wohlfarth theory of itinerant ferromagnetism. However, low-temperature specific-heat data[1,3] are inconsistent with the Stoner-Wohlfarth model. Furthermore, Chu *et al.*[4] have found the Curie temperature (T_C) to be unshifted by the application of hydrostatic pressure up to 18 kbar, whereas the band model predicts a depression of T_C under pressure. Robbins and Claus[3] have interpreted their magnetization and low-temperature specific-heat data by postulating the formation of large magnetic clusters. However, this hypothesis appears to be inconsistent with the existence of the sharp paramagnetic-to-ferromagnetic transition at T_C.

Chang *et al.*[5] have measured the magnetoresistance of stoichiometric and off-stoichiometric polycrystalline samples at 4.2 K in fields up to 340 kOe and have developed a simple two-band model for the magnetoresistance of an itinerant ferromagnet. In paramagnetic samples they obtained good agreement between theory and experiment. However, in ferromagnetic samples the agreement was good only at high fields ($H > 100$ kOe) where the negative magnetoresistance arising from magnetic effects was masked by the "conventional" positive magnetoresistance arising from the orbital motion of the conduction electrons in a magnetic field.

In this paper we report transverse magnetoresistance measurements[6] over the temperature range $1.2 < T < 90$ K in fields up to 23 kOe. Due to the relatively low magnetic fields and high temperatures, our data are dominated by the negative magnetoresistance, allowing a better investigation of the influence of magnetic ordering. Our data are in qualitative agreement with the predictions of the theory of Chang *et al.* However, the observed magnetoresistance is larger than predicted by a factor of 2–20 depending on composition and temperature.

II. EXPERIMENTAL TECHNIQUE

A series of ingots with Ni concentrations ranging from 74 to 76 at. % was prepared by arc melting followed by vacuum annealing at 1000 °C for 21 days and water quenching.[7] Polycrystalline samples were cut from these ingots with an abrasive wheel. Potential leads were spot-welded to the samples while current leads were either spot-welded or soldered. Figure 1 shows the residual-resistance ratio ($\rho_{300\,K}/\rho_{4.2\,K}$) as a function of Ni concentration, exhibiting a peak at the stoichiometric composition as expected for a good intermetallic compound.

Curie temperatures were determined by low-frequency ($f = 20$ Hz) ac susceptibility measurements. A small pickup coil of 10–20 turns was wound around each magnetoresistance sample so these measurements could be made without re-

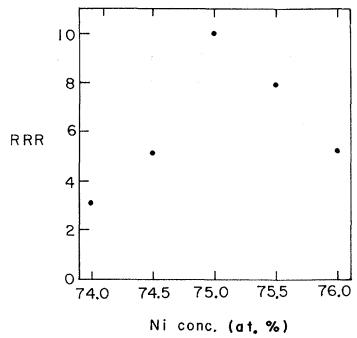

FIG. 1. Residual-resistance ratio (RRR) vs Ni concentration.

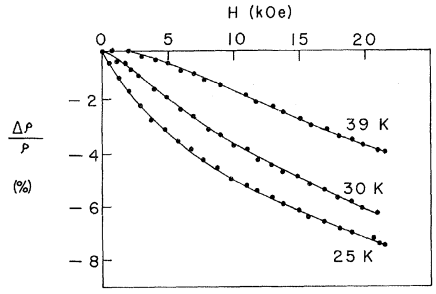

FIG. 2. Magnetoresistance vs magnetic field for a nominally stoichiometric sample (T_C = 25 K).

mounting. The Curie temperatures of our samples are compared with those of De Boer *et al.*[2] in Table I. The discrepancies presumably are due to slight differences in composition or strain.

Magnetoresistance measurements were made by sweeping the magnetic field while the temperature was held constant by an exchange gas system. The possibility of errors arising from pickup of the Hall voltage was checked by rotating the magnetic field in the plane normal to the current direction. Errors from this source are less than 0.02% of the zero-field resistivity for the 74.0-at. %-Ni sample and less than 0.1% for the other samples.

III. RESULTS

Negative magnetoresistance was observed in all cases. For example, the field dependence of the magnetoresistance of a nominally stoichiometric sample is shown in Fig. 2 for several temperatures. The magnetoresistance is normalized in the usual way, i.e.,

$$\Delta\rho/\rho \equiv [\rho(H, T) - \rho(0, T)]/\rho(0, T).$$

The temperature dependence of the magnetoresistance measured at H = 21 kOe is shown in Fig. 3 for the five compositions studied. The magnetoresistance of the ferromagnetic samples exhibits a pronounced peak at intermediate temperatures; this peak coincides closely with T_C for Ni concentrations of 75 at. % or greater but occurs at a somewhat higher temperature in the 74.5-at. %-Ni sample.

TABLE I. Curie temperatures of the samples.

Ni conc. (at. %)	T_C (K) Our samples	De Boer *et al.*[a]
74.0	b	c
74.5	2	c
75.0	25	41.5
75.5	55	58.1
76.0	67	71.5

[a]Reference 2.
[b]Paramagnetic down to 1.2 K.
[c]Paramagnetic down to 4.2 K.

TABLE II. Comparison of our magnetoresistance data for H = 21 kOe with the theory of Chang *et al.*

Ni conc. (at. %)	T (K)	$\Delta\rho/\rho$ (%) Experiment	Theory
74.0	4.2	−0.3	−0.05
74.0	20	−0.2	−0.05
75.0	4.2	−1	−0.4
75.5	4.2	−2.1	−0.5
75.5	77	−1.6	−0.1

Our results at T = 4.2 K are qualitatively in agreement with those of Chang *et al.* for corresponding compositions and magnetic field strengths except for nearly stoichiometric samples; their samples containing 74.7-at. % and 74.8-at. % Ni showed positive magnetoresistance at fields larger than 20 kOe. This is further evidence that our nominally stoichiometric sample may in fact be slightly Al rich, since even a slight departure from stoichiometry would increase the electron scattering rates and noticeably diminish the positive magnetoresistance effect.

IV. DISCUSSION

In the two-band model of Chang *et al.*,[5] the current flow is dominated by s-band electrons which

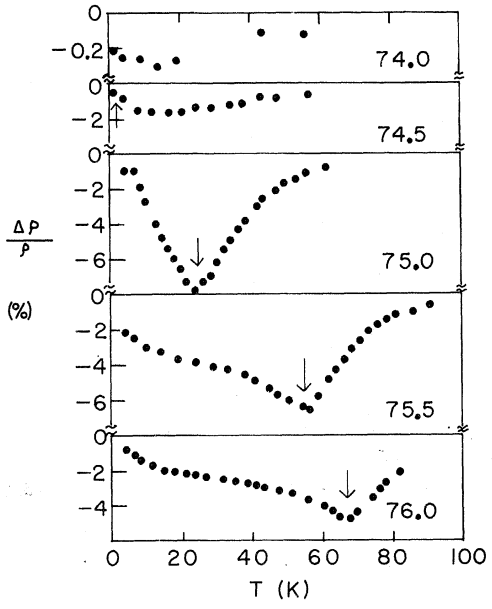

FIG. 3. Magnetoresistance at H = 21 kOe vs temperature for five different compositions. Note different scale for topmost curve. Arrows indicate Curie temperatures determined from ac susceptibility.

may be scattered to other s states or to d states by impurity atoms, defects, or phonons. In the presence of bulk magnetization (either spontaneous or induced by an external field) the d-band densities of states at the Fermi energy for spin-up and spin-down electrons are unequal. This disparity, which can be determined from magnetization measurements, causes the scattering rates for spin-up and spin-down s-band electrons to differ, giving rise to a negative term in the magnetoresistance. The observed magnetoresistance represents a combination of this negative effect and the conventional positive magnetoresistance effect. Chang *et al.* have empirically adjusted the band parameters and Kohler function in order to fit their magnetoresistance data taken at 4.2 K in fields up to 340 kOe.

We have calculated the magnetoresistance of some of our samples at various temperatures using the theory of Chang *et al.*[5] with their empirical values of band parameters and Kohler function, the magnetization data of De Boer *et al.*,[2] and the zero-field resistivities of our particular samples. The theoretical values are compared with our data in Table II. In all cases the theoretical values are much too small. Unfortunately we cannot calculate the magnetoresistance of the ferromagnetic samples at intermediate temperatures due to lack of magnetization data. However, by making a rough estimate from the data of De Boer *et al.* we find that the theory predicts a peak in $\Delta\rho/\rho$ at $T \simeq T_C$ but that the magnitude of the peak is too samll by at least a factor of 5. It should be noted that our use of the magnetization data of De Boer *et al.* rather than data from our own samples could introduce some error into the theoretical predictions of $\Delta\rho/\rho$, since our samples may differ slightly in composition from theirs. However, it is unlikely that our Ni concentrations are in error by more than 0.1 at. % except possibly in the nominally stoichiometric sample. Since we estimate that an error of this magnitude would change the theoretical $\Delta\rho/\rho$ by less than $\frac{1}{3}$ of the predicted value, such an error is not significant except perhaps for the nominally stoichiometric sample.

Taking our data together with those of Chang *et al.* we find that for ferromagnetic samples the agreement between theory and experiment is good only for data taken at high fields and low temperatures,[5] where the magnetoresistance is strongly positive. In that case the "orbital" effect predominates over the magnetic effect; thus the theoretical values depend mainly on the Kohler function and are insensitive to the magnitude of the magnetization and the choice of band parameters. Agreement is poor for data taken at low fields and higher temperatures, where the magnetoresistance is negative. In this case the magnetic effect predominates over the "orbital" effect; here the theoretical values depend mainly on the magnetization and band parameters rather than the Kohler function.

It therefore appears that the empirical Kohler function used by Chang *et al.* is correct and that the difficulty lies in their treatment of the magnetic effect. Agreement between theory and experiment could be improved by drastic modification of the empirical band parameters although the question of their physical significance would then arise. However, we wish to point out that negative magnetoresistance with a temperature dependence similar to that shown in the lower three curves of Fig. 3, including a peak at $T = T_C$, is observed in the localized-moment ferromagnets $CdCr_2Se_4$ (Ref. 8) and *Pd*-Fe (Ref. 9) and is believed to be due to field-induced suppression of spin-disorder scattering.

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