Residual resistivity in Ni-Fe-Cr/V alloys and the two-current model

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Abstract. The resistivity of a large number of Ni-Fe-Cr/V alloys has been measured at 77 K. An attempt has been made to explain the concentration dependence of resistivity in the light of Mott's two-current model. It is seen that contrary to earlier observation, the model is applicable to a larger range of concentration for the Ni-Fe-Cr series. Appreciable deviation from the model starts for Fe concentration exceeding about 15 at. % and Cr about 7 to 8 at. %. However, for similar concentration range in Ni-Fe-V series, disagreement between the experimental and theoretical results is more pronounced.

Keywords. Resistivity; ternary alloys; two-current model.

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1. Introduction

Recently we have reported the extraordinary Hall effect (Gangopadhyay et al. 1984a) and detailed magnetization measurements (Gangopadhyay et al. 1984b) on a series of nickel-rich Ni$_{1-x-y}$Fe$_x$Cr$_y$ and Ni$_{1-x-y}$Fe$_x$V$_y$ alloys. Although the resistivity data for these alloy series were also reported (Gangopadhyay et al. 1984a), no attempt was made to explain the change in resistivity with alloy concentration. In this paper we have tried to explain the resistivity in terms of the "two-current model." It should, however, be mentioned that the applicability of the two-current model has been tested earlier (Dorleijn 1976) in the dilute alloy limit (impurity concentration < 5 at. %). We have tried to check whether it can also be applied to fairly concentrated alloys (impurity concentration in the range 15 to 30 at. %).

To explain the anomalous temperature dependence of resistivity in Ni and Pd, Mott (1936) suggested the two-current model according to which electronic conduction takes place in parallel, through spin-up and spin-down electrons in transition metals. The idea was developed by Fert and Campbell (1968) and later extended by Leonard et al. (1969), Dorleijn (1976) and others. The basic philosophy behind this model is that in transition metals, the relaxation times of the spin-up ($S \uparrow$) and spin-down ($S \downarrow$) electrons are quite different and they conduct in parallel. The observed large deviation from Mathiessen's rule (DMR) in transition metal alloys lends strong support to this hypothesis. Different band occupancies of the $d$-electrons, which are split into $S \uparrow$ and $S \downarrow$ bands in the presence of exchange field, provide the main reason for the different relaxation times for electrons with opposite spins. Basically the mechanisms responsible for resistivity in transition metals are: (i) the direct $s$-$s$ scattering as in normal
metals, (ii) the indirect s-d-s scattering, and (iii) the s-d scattering. If in a particular metal like Ni, the S \uparrow band is full, the resistivity contribution of the S \uparrow electrons ($\rho \uparrow$) will be considerably less than $\rho \downarrow$ since the third process mentioned above cannot take place for the S \uparrow electrons.

During the last 15 years considerable data in terms of the two-current model have been published on resistivity (Dorleijn 1976 and references cited therein) of transition metals and alloys, thermoelectric power in various Ni and Co-based transition metal alloys belonging to the 3d, 4d and 5d series (Cadeville and Roussel 1971), ferromagnetic anisotropy of resistance and extraordinary Hall effect in dilute Ni and Fe-based transition metal alloys (Dorleijn 1976; Dorleijn and Miedema 1976, 1979). Also the so-called “period effect,” which is characterised by: (i) a maximum in $\rho \uparrow$ (Durand and Gautier 1970), (ii) sign change of the thermoelectric power (Cadeville and Roussel 1971) and (iii) a maximum in the specific increment of the electronic specific heat coefficient (Caudron et al 1973), finds suitable explanation in the framework of the two-current model. This “period effect” is observed when various elements, starting from the beginning of the transition metal series are added into Ni (or some other element of the series) and the changes in the various physical properties mentioned earlier, are monitored. A distinct anomaly is observed when Ni is diluted with Cr of the first transition series, Ru of the second and Os of the third series. This is attributed to the coincidence of the virtual-bound state of the impurity atom with the Fermi-level of the host element.

Using such a well-tested model to our resistivity data, we have observed that within the limitations of the model, our results agree with the model predictions for alloys in the Cr-series reasonably well. However, considerable deviations have been observed for the alloys in the vanadium series.

2. Experimental

Since the details of the alloy preparation and measurement techniques were reported earlier (Gangopadhyay et al 1984a), we briefly discuss only the experimental procedure. The composition of the alloys was chosen mainly as a function of Cr/V concentration and in some cases as a function of increasing Fe concentration. Induction melt ingots were homogenized at 1050°C for 48 hr, quenched in water and finally annealed (900°C, 24 hr) and again quenched in water (to retain the disordered structure) after various metallurgical processes. The alloy compositions were analyzed using atomic absorption spectrometry. Resistivity was measured using the usual four-probe d.c. method.

3. Results and discussion

In table 1 are presented the 77 K resistivity data for some Ni-Fe-Cr/V alloys. The error bars shown in the data (inclusive of all possible factors) mainly come from the dimension measurements of the samples of which the lowest dimension i.e. the thickness contributes the maximum. Since all the alloys considered are fairly concentrated, impurity contribution to resistivity dominates all other effects, e.g. thermal contribution etc. Hence the change in resistivity between 77 K and 4 K will be
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Table 1. Experimental values of the resistivity of some Ni-Fe-Cr/V alloys at 77 K and those predicted by the two-current model (equation (3)).

<table>
<thead>
<tr>
<th>Alloy type</th>
<th>Alloy composition (at. %)</th>
<th>( \rho_{\text{expt}} (\mu \Omega \cdot m) ) at 77 K</th>
<th>( (\rho_{\text{th}})^{\dagger} ) residual (( \mu \Omega \cdot m ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni-Fe-Cr</td>
<td>76.8 - 21-2 - 2</td>
<td>0.43 ± 0.01 (0.53)*</td>
<td>0.41</td>
</tr>
<tr>
<td></td>
<td>85.5 - 11 - 3.5</td>
<td>0.52 ± 0.01 (0.58)</td>
<td>0.43</td>
</tr>
<tr>
<td></td>
<td>80 - 16 - 4</td>
<td>0.63 ± 0.03 (0.69)</td>
<td>0.56</td>
</tr>
<tr>
<td></td>
<td>75.5 - 20 - 3 - 4.2</td>
<td>0.61 ± 0.02 (0.70)</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>69.6 - 22 - 8 - 7.6</td>
<td>0.90 ± 0.01 (0.96)</td>
<td>0.92</td>
</tr>
<tr>
<td>Ni-Fe-V</td>
<td>75 - 17 - 8</td>
<td>0.77 ± 0.02 (0.83)</td>
<td>0.84</td>
</tr>
<tr>
<td></td>
<td>67 - 21 - 12</td>
<td>1.04 ± 0.03 (1.10)</td>
<td>1.17</td>
</tr>
<tr>
<td></td>
<td>75.1 - 12.8 - 12.1</td>
<td>0.93 ± 0.02</td>
<td>0.98</td>
</tr>
<tr>
<td></td>
<td>72.5 - 13.7 - 13.8</td>
<td>0.99 ± 0.02</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td>68.1 - 17.4 - 14.5</td>
<td>1.04 ± 0.05</td>
<td>1.23</td>
</tr>
<tr>
<td></td>
<td>73.5 - 15.5 - 15</td>
<td>1.03 ± 0.04</td>
<td>1.17</td>
</tr>
<tr>
<td></td>
<td>78 - 6 - 16</td>
<td>0.84 ± 0.04</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td>70 - 12 - 18</td>
<td>1.06 ± 0.03 (1.10)</td>
<td>1.27</td>
</tr>
<tr>
<td></td>
<td>72 - 8 - 20</td>
<td>1.07 ± 0.03 (1.11)</td>
<td>1.26</td>
</tr>
<tr>
<td>Ni-Fe-V</td>
<td>81 - 17 - 2</td>
<td>0.34 ± 0.01 (0.44)</td>
<td>0.26</td>
</tr>
<tr>
<td></td>
<td>79.4 - 17 - 3.6</td>
<td>0.49 ± 0.01 (0.60)</td>
<td>0.37</td>
</tr>
<tr>
<td></td>
<td>85 - 11 - 4</td>
<td>0.54 ± 0.02</td>
<td>0.35</td>
</tr>
<tr>
<td></td>
<td>74 - 22 - 4</td>
<td>0.58 ± 0.02</td>
<td>0.44</td>
</tr>
<tr>
<td></td>
<td>83 - 10 - 7</td>
<td>0.81 ± 0.02 (0.90)</td>
<td>0.49</td>
</tr>
</tbody>
</table>

* Numbers in parentheses represent the corresponding quantities at 300 K.

† The uncertainty in the calculated values, originating from the determination of the actual alloy composition, is estimated to be less than 5%.

rather small, at least for the concentrated alloys, as could be estimated from the change in resistivity between room temperature and 77 K. To give some idea about this expected change in \( \rho \), the corresponding 300 K data for some of the representative alloys are shown in table 1. It can be seen that at least for the concentrated alloys \( \rho (77 \text{K}) \) can be considered to be very close to \( \rho (4 \text{K}) \), within the error limits of our data. However, for lower concentrations, \( \rho (77 \text{K}) \) can exceed \( \rho (4 \text{K}) \) by a maximum of about 7–8%. Within this limitation, \( \rho (77 \text{K}) \) can therefore be taken as representative of the residual resistivity \([\rho (4 \text{K})]\) of the alloys. Table 1 also presents the theoretically calculated residual resistivity of the alloys according to the two-current model. The method of calculation is discussed below.

According to the two-current model, the total electrical resistivity \( \rho \), is given by

\[
\rho = \frac{\rho_T \rho_I + \rho_{II}(\rho_I + \rho_{II})}{\rho_I + \rho_{II} + 4\rho_{II}},
\]

where \( \rho_{\sigma}(\sigma = 1, 1) \) are the resistivities associated with each sub-band \( \sigma \), and \( \rho_{II} \) is the term which takes care of the mixing of the two parallel currents due to spin-flip scattering (electron-magnon interaction) and is important only at fairly high temperatures. It is assumed that although Matthiessen’s rule is not valid for the total resistivity, each sub-band resistivity follows the same. Thus in a binary alloy \( A_{100-x}B_x \), the sub-
band resistivities are given by
\[ \rho_e(T) = \rho_{m0}(T) + x\rho_{io}, \] (2)
where the first term on the right side signifies the pure matrix resistivity and the second term, the impurity resistivity which is assumed to be temperature-independent. It should also be noted that in the linear relationship between impurity resistivity and concentration \( x \), is the inherent implication that (2) should be valid in the dilute alloy limit where the impurity-impurity interaction is insignificant. Experimental results (Dorleijn 1976) show that (2) is valid up to about 5 at. % of total impurity content in most of the Ni- and Fe-based alloys.

Now in the dilute alloy limit and at low temperatures, the pure matrix resistivity term \( \rho_{m0} \) and the spin-mixing term \( \rho_{io} \) are insignificant compared to the impurity resistivity \( \rho_{io} \). Under this condition, combining (1) and (2), the expression for the residual resistivity is given by
\[ \rho_0 = \frac{x\rho_{io} + \rho_{io}}{\rho_{io} + \rho_{io}}. \] (3)
In case of a ternary alloy \( A_{100-x-y}B_xC_y \), in the low temperature limit, \( \rho_e \) is given by
\[ \rho_{io} = x\rho_{Bo} + y\rho_{Co}. \] (4)
Therefore, if \( \rho_{Bo}, \rho_{Co} \) are known, the residual resistivities can be easily calculated. Studying hundreds of binary and ternary alloys based on Ni and Fe, Dorleijn (1976) tabulated the values of \( \rho_e \) for different transition metal impurities. Using the values of \( \rho_e \) for Fe, Cr, V etc in Ni, one could estimate the residual resistivities and compare with our experimental values. But one should not expect very good agreement with (3) simply because our alloys are fairly concentrated and due to impurity-impurity interactions, the linear relationship between impurity resistivity and concentration (4) cannot be expected to hold good. According to Dorleijn (1976) \( \rho_{Fe} = 0.41, \rho_{Fe} = 49 \mu\Omega\text{cm}; \rho_{Cr} = 28 \) and \( \rho_{Cr} = 6.2 \mu\Omega\text{cm}, \rho_{V} = 14 \) and \( \rho_{V} = 6.5 \mu\Omega\text{cm} \) respectively. Using these values in (3) and (4), resistivity of our alloys was calculated and presented in table 1. One should note that because of the experimental error involved in determining the actual composition of the sample, the calculated values are reliable within \( \pm 5 \% \) (Gangopadhyay et al 1984a).

Resistivity for Ni-Fe-Cr alloys is presented in two different groups in table 1. The first group consists of those alloys having less than about 8 at. % of Cr and the rest greater than that. It can be seen that for the first group of alloys, if \( \rho(77 K) \) is normalized to \( \rho(4 K) \) using the observed change between 300 K and 77 K, the theoretical and experimental values are well in agreement if the error bars of the experimentally measured quantities and the inherent limitations of the "two-current model" are kept in mind. The theoretical values are either in agreement (within error limits) or slightly higher than the experimental ones, except for the alloy (85.5-11-3.5). But for the second group of alloys the agreement is poorer and deviations are greater for the alloys richer in Cr with theoretical values exceeding (by a maximum of about 20 % or so) the experimental ones. In the context of the foregoing discussions, reasonable agreement with the model even for the first group of alloys is also surprising since the model is supposed to work for total impurity concentrations not exceeding 5 at. %. We attribute the extended range of agreement to the following reasons:

(i) Theoretical calculations (Akai 1977) based on the coherent potential approxima-
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section for Ni-Fe and Ni-Cr alloys show that in Ni-Fe alloys, \( \rho_{Fe1} \) is approximately linear even up to about 15 at. % of Fe, after which it starts bending and ultimately decreases after passing through a maximum around 30 at. % of Fe. But \( \rho_{Fe1} \) remains very small until about 60 at. % of Fe. Hence deviation from (4) should not be much even up to 15 at. % of Fe as we have observed.

(ii) Since the contribution of Cr atoms to the total resistivity is much larger than that of Fe atoms (\( \rho_{Cr1} \gg \rho_{Fe1} \) and \( \rho_{Cr1} \sim \rho_{Fe1} \)), in ternary Ni-Fe-Cr alloys the effect of small non-linearity in \( \rho_{Fe} \) should not very much influence the total resistivity. Hence, even for alloys with high Fe but low Cr contents, disagreement between theoretical and experimental values should not be appreciable and is consistent with our observations in table 1.

But in the case of alloys with high Cr content (> 7 to 8 at. %), theoretical values are always larger than the experimental ones. The reason is that theoretical (Akai 1977) as well as experimental results (Dorleijn 1976) have pointed out that in binary Ni-Cr alloys, resistivity increases linearly with concentration only up to about 5 to 6 at. % of Cr and then the rate of increase slows down. So if one calculates \( \rho \) for the concentrated alloys, using the values of \( \rho_{Cr1} \) and \( \rho_{Cr1} \) in the linear range (C < 5 at. %), it is quite obvious that the calculated values will be larger than the actual ones. Since the resistivity is dominated by Fe content, disagreement between theoretical and experimental values will be greater for the alloys richer in Cr-content. This is consistent with our observation.

However, the same type of calculations, when extended to Ni-Fe-V alloys, hardly met with any success. Even for the alloys with low V content, the disagreements were enormously large, as can be seen from table 1. Since for the alloys with higher V-content, the disagreements were even larger, they are not included in the table. One interesting observation from table 1 is that the calculated values are always much smaller than the observed ones. As discussed earlier, if deviation from linearity of the \( \rho \) vs concentration curves [i.e. violation of (4)] would have been the reason, one should have expected the calculated values to be larger than the observed ones. Again as \( \rho_{V1} \) < \( \rho_{Cr1} \) and \( \rho_{V1} \sim \rho_{Cr1} \), one should expect that the ternary Ni-Fe-V alloys should have lower resistivities compared to the same compositions for the corresponding Ni-Fe-Cr alloys. But surprisingly, we observe from our experimental result that \( \rho \) for the alloys in both the series with similar compositions, has comparable magnitudes.

4. Conclusion

From the present analysis we conclude that: (i) the two-current model can fairly well explain the residual resistivity of Ni-Fe-Cr system even for an extended range of impurity concentration. Appreciable deviation from the model starts for Cr concentration exceeding about 7 at. %, whereas for Fe even at about 20 at. %, disagreement is not large. However, for Ni-Fe-V system the model fails to explain the resistivity behaviour in the range of concentration studied by us.

It is quite possible that since V is one step away from Ni in the periodic table than Cr, chances of a separate band/sub-band formation, distinct in energy scale from the bands of the host metal (termed as “split bands”, the details of which can be found in Gangopadhyay et al 1984a), is more in the case of vanadium alloys than the corresponding Cr ones. In such an event the applicability of the two-current model
(where change in band structure on alloying is neglected) will be seriously limited and probably our experimental results point to this direction. In fact our earlier Hall effect measurements (Gangopadhyay et al 1984a) also pointed out that vanadium alloys correspond more to the split-band description than the Cr ones. So it appears that the resistivity data, together with the earlier Hall effect data, indicate that ternary vanadium alloys fit into the split-band picture better; whereas the band structure of corresponding Cr alloys lies somewhere between the rigid-band and the split-band models. For a clear understanding of the systems, once again the present work emphasizes the need for a theoretical band structure calculation of the ternary alloys under consideration.

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