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TRANSPORT PROPERTIES OF FERROMAGNETIC TRANSITION METALS

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Résumé. — Nous passons en revue des expériences récentes montrant que la conduction dans les métaux ferromagnétiques de transition à basse température est par deux courants en parallèle d'électrons de spin ↑ et de spin ↓, mélangés par les collisions avec retournement du spin quand la température s'élève. Les valeurs relatives des résistivités d'impuretés de métaux de transition pour les électrons de spin ↑ et de spin ↓ sont expliquées. Un modèle à deux courants permet de comprendre plus précisément de nombreuses autres propriétés de transport comme l'anisotropie spontanée de la résistivité, le pouvoir thermoélectrique, la conductivité thermique, l'effet Hall ordinaire et extraordinaire, la magnétorésistance. Quelques commentaires sur la résistivité près du point de Curie sont aussi présentés.

Abstract. — We review recent experiments showing that electrical conduction in transition ferromagnetics at low temperature is by two currents of spin ↑ and spin ↓ electrons in parallel, mixed by spin-non-conserving collisions when temperature increases. The relative values of transition impurity resistivities for spin ↑ and spin ↓ electrons are explained. A two current model allows a better understanding of many other transport properties, such as spontaneous anisotropy of the resistivity, thermoelectric power, thermal conductivity, ordinary and extraordinary Hall effect, magnetoresistance. Some comments on the resistivity near the Curie point are also presented.

We will summarize briefly recent work on transport properties of transition ferromagnetics, concentrating particularly on effects that are specific to their ferromagnetic character.

I. Resistivity : general. — Electrons in a given domain of a ferromagnet can be strictly classified as having spin up (i. e. parallel to the majority spins) or down, if the spin-orbit coupling is ignored and the spin-flip transitions supposed not too frequent. Hence, the conduction is by two currents in parallel with different resistivities, this difference resulting mainly from the difference in the possibilities of s-d scattering for the s electrons.

1) In the low temperature limit, as only scattering with conservation of spin direction is allowed (*), the conduction is by two independent currents. Hence the residual resistivity ρ_0 is given by

$$\frac{1}{\rho_0} = \frac{1}{\rho_{0\uparrow}} + \frac{1}{\rho_{0\downarrow}}, \text{ i. e. } \rho_0 = \frac{\rho_{0\uparrow} \rho_{0\downarrow}}{\rho_{0\uparrow} + \rho_{0\downarrow}} \quad (1)$$

where $\rho_{0\uparrow}$ and $\rho_{0\downarrow}$ are the residual resistivities for the spin ↑ and spin ↓ electrons.

The residual resistivity of a ternary dilute alloy MAB is expected to be

$$\rho_0(\underline{\text{MAB}}) = \frac{(\rho_{A\uparrow} + \rho_{B\uparrow})(\rho_{A\downarrow} + \rho_{B\downarrow})}{\rho_{A\uparrow} + \rho_{B\uparrow} + \rho_{A\downarrow} + \rho_{B\downarrow}}$$

which means a deviation from Matthiessens' rule :

$$\Delta\rho = \rho_0(\underline{\text{MAB}}) - \rho_0(\underline{\text{MA}}) - \rho_0(\underline{\text{MB}}) = \frac{(\alpha_A - \alpha_B)^2 \rho_0(\underline{\text{MA}}) \rho_0(\underline{\text{MB}})}{(1 + \alpha_A)^2 \alpha_B \rho_0(\underline{\text{MA}}) + (1 + \alpha_B)^2 \alpha_A \rho_0(\underline{\text{MB}})} \quad (2)$$

where $\alpha_A = \rho_{A\downarrow}/\rho_{A\uparrow}$ and $\alpha_B = \rho_{B\downarrow}/\rho_{B\uparrow}$. Measurements [1, 2, 3, 4] of residual resistivities of iron or nickel containing various pairs of impurities show very large deviations from Matthiessens' rule

(*) For residual scattering by impurities, it is known [5], that spin-flip scattering is generally small compared to scattering without spin-flip.

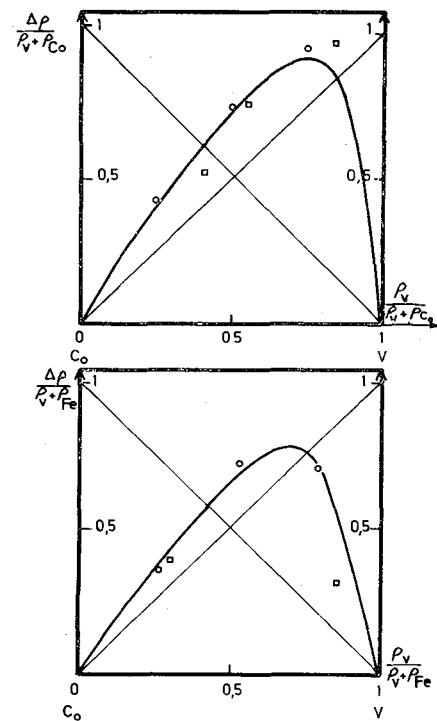


FIG. 1. — Relative deviations from Matthiessen's rule for NiCoV and NiFeV alloys [3]. The full curve indicates the deviation calculated from expression (2) with $\alpha_{Co} = 30$, $\alpha_{Fe} = 20$, $\alpha_V = 0.65$.

(Fig. 1) corresponding to a considerable dispersion of the α values and allow an experimental determination of these values (exemple : in nickel [2], $\alpha_{Co} \approx 30$, $\alpha_{Cr} \approx 0.4$, ...)

2) As the temperature is increased, a thermal contribution must be added to each resistivity ;

$$\rho_\sigma = \rho_{0\sigma} + \rho_\sigma^i(T)$$

but also spin-non-conserving scattering will occur in electron-magnon or electron-electron collisions and mix the two currents. We can write coupled Boltzmann equations for the two directions of spin, which

is roughly equivalent in a classical model to writing the following coupled equations for the average velocities v_σ

$$-\frac{eE}{m} = \frac{v_\uparrow}{\tau_\uparrow} + \frac{v_\uparrow - v_\downarrow}{\tau_{\uparrow\downarrow}}$$

$$-\frac{eE}{m} = \frac{v_\downarrow}{\tau_\downarrow} + \frac{v_\downarrow - v_\uparrow}{\tau_{\uparrow\downarrow}}$$

where $\tau_{\uparrow\downarrow}$ is the relaxation time characteristic of the momentum transfer between the two currents. One finds [1, 6]:

$$\rho = \frac{\rho_\uparrow \rho_\downarrow + \rho_{\uparrow\downarrow}(\rho_\uparrow + \rho_\downarrow)}{\rho_\uparrow + \rho_\downarrow + 4\rho_{\uparrow\downarrow}} \quad (3)$$

the resistivity ρ depending on the resistivities ρ_\uparrow , ρ_\downarrow and in the case where $\rho_\uparrow \neq \rho_\downarrow$ on a mixing term $\rho_{\uparrow\downarrow}$. $\rho_{\uparrow\downarrow}$ expresses the strength of the spin \downarrow -spin \uparrow mixing processes (*) and normally increases with temperature.

The dependance of the resistivity on $\rho_{\uparrow\downarrow}$ expressed in (3) can be described as follows: suppose, for example, an alloy where residual resistivities $\rho_{0\uparrow}$ and $\rho_{0\downarrow}$ are very different; at low temperature, one current path will short circuit the other; when the temperature increases, the fast current will be « braked » by mixing with the slow current and the resistance will increase much faster than in the pure metal, leading to large deviations from Matthiessen's rule.

These deviations have been observed between 1 °K and 300 °K in Fe [1, 3], Ni [2] [3, 7, 8] and Co [9] dilute alloys with transition metals impurities. Deviations can be very large: for example, in NiCo alloy ($\rho_{\text{Co}\downarrow}/\rho_{\text{Co}\uparrow} \approx 30$), the variation of the resistivity below 30 °K is more than four times the variation in pure nickel and the apparent resistivity of Co impurities at 300 °K is about three times their residual resistivity; in absolute terms, deviations measured can easily be $\sim 5 \mu\Omega \text{ cm}$ compared with values $\sim 10^{-2} \mu\Omega \text{ cm}$ for deviations observed in ordinary metals [10].

Analysis of these temperature deviations gives values of $\rho_{0\uparrow}$, $\rho_{0\downarrow}$ for transition metals impurities in agreement with those deduced from ternary alloy measurements and in addition gives values of $\rho_\uparrow^i(T)$, $\rho_\downarrow^i(T)$, $\rho_{\uparrow\downarrow}^i(T)$; within the accuracy of experiments these temperature dependant resistivities are independent of the impurity present, and so they are « pure metal » parameters.

II. Experimental residual resistivities. — As an example of the data extracted, values of $\rho_{0\uparrow}$, $\rho_{0\downarrow}$ for impurities of the first transition series metals in nickel are shown in figure 2.

These results provide a striking justification of Friedel's virtual bound state model [11].

Thus, for systems such as NiCo, NiFe, NiMn, lying on the Slater-Pauling curve, all the shielding

(*) Note that the spin-non-conserving processes contribute also to ρ_\uparrow and ρ_\downarrow ; for example, spin-flip scattering between $\mathbf{k}\uparrow$ and $\mathbf{k}\downarrow$ free electron states gives a contribution proportional to $|\langle \mathbf{k} - \mathbf{k}' \cdot \mathbf{u} \rangle|^2$ for ρ_\uparrow or ρ_\downarrow , and proportional to $\langle \mathbf{k}, \mathbf{u} \rangle \langle \mathbf{k}', \mathbf{u} \rangle$ for $\rho_{\uparrow\downarrow}$ (\mathbf{u} is the unitary vector along the electric field).

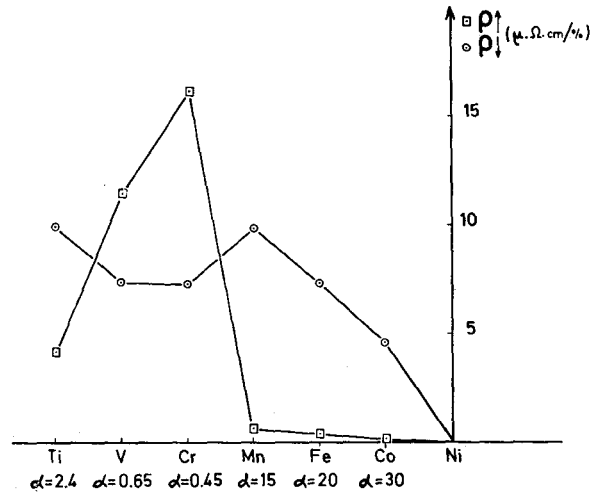


FIG. 2. — Experimental residual resistivities $\rho_{0\uparrow}$, $\rho_{0\downarrow}$ for first transition series impurities in nickel [3].

of the impurity is done by the $d\uparrow$ electrons, the $d\downarrow$ states remaining below the Fermi level; hence $\rho_{0\uparrow}$, due only to s-s scattering, is much smaller than $\rho_{0\downarrow}$.

For NiCr, $d\uparrow$ bound states are repelled above the Fermi level; this leads to the negative magnetic moment of the Cr impurity and to the peak in $\rho_{0\uparrow}$, due to the formation of a virtual bound state at the Fermi surface in the $s\uparrow$ band. The effect is analogous to the well known residual resistance behaviour for transition impurities in noble metals.

Similar effects have been obtained for first transition series impurities in iron [3] and cobalt [9]. Gautier and coworkers [8, 9] have also shown that there are marked series effects for heavier transition impurities in Ni and Co, the virtual bound state in the spin \uparrow band appearing for a smaller impurity charge difference in the second and third series. These effects have been explained on a simple model by Demangeat and Gautier [12].

III. Host resistivity parameters. — For the temperature dependant parts of ρ_\uparrow and ρ_\downarrow , the ratio $\rho_\downarrow^i(T)/\rho_\uparrow^i(T)$ varies for Ni [2, 3] between 3 at low temperature and 5 at 100 °K, and for Co [9] from 23 to 13 in the same limits. These values are in agreement with the s-d scattering model: spin \downarrow conduction electrons can be scattered into $d\downarrow$ states at the Fermi surface by electron-phonon or electron-electron collisions, which gives a much higher $\rho_\downarrow^i(T)$ than $\rho_\uparrow^i(T)$; it appears that magnon-electron collisions do not contribute much to $\rho_\uparrow^i(T)$ or $\rho_\downarrow^i(T)$ (but they contribute to the final resistivity via $\rho_{\uparrow\downarrow}^i$).

The spin mixing term $\rho_{\uparrow\downarrow}^i(T)$ can be shown from its importance to be due to magnon-electron scattering [6] rather than to electron-electron collisions [13]. Its experimental variation with temperature (Fig. 3 for Ni) is in agreement with the variation predicted by simple models [3, 13] for scattering by magnons; the dip in $\rho_{\uparrow\downarrow}^i/T^2$ below 25 °K can be ascribed to the separation in k space between spin \uparrow and spin \downarrow conduction electrons Fermi surfaces: when the q of the magnon can no longer span this gap, the number of

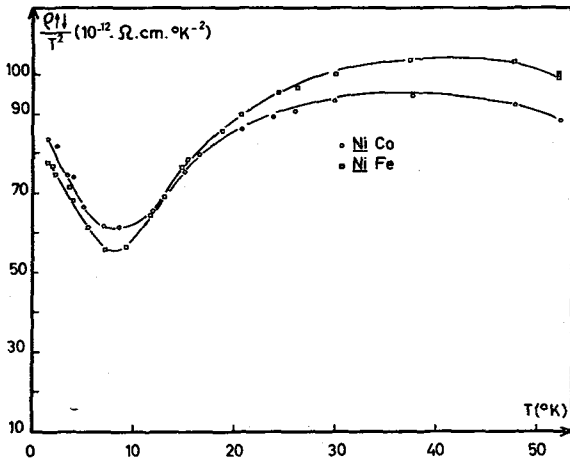


FIG. 3. — Experimental variation of ρ_{\parallel}/T^2 with T . Circles and squares are values deduced from measurement on NiCo or NiFe alloys.

possible scattering processes will drop exponentially. For $T \lesssim 10$ °K, the behavior is more complicated, ρ_{\parallel} varying as $T^{3/2}$ (shown by the upturn in the ρ_{\parallel}/T^2 plot) and depending on impurity concentration. This appears to have the same origin as the $T^{3/2}$ term found by Williams [15] and Turner and Long [14] for dilute PdFe and PdCo. In impure ferromagnetics, when the magnon wavelength is greater than the distance between impurities, collisions become incoherent and magnon-electron scattering no longer needs to conserve total momentum. The scattering rate will then depend only on the number of magnons, and so is proportional to $T^{3/2}$.

IV. Spontaneous resistivity anisotropy. — It is well known that for a ferromagnet at technical saturation, the resistivity is different for a current parallel or perpendicular to the magnetization direction, and it has been realized for a long time that this is due to a spin-orbit effect. We have shown [16, 17] that the model of Smit [18] outlined below taken with the two current resistivity data gives an excellent explanation of the resistivity anisotropy in Ni based alloys.

The $d\downarrow$ electrons at the Fermi surface have some $d\uparrow$ character mixed in by the spin-orbit interaction, and a little of the $d\downarrow$ character is mixed out; the spin \uparrow conduction electrons can then be scattered into the $d\uparrow$ part of the $d\downarrow$ Fermi surface, and there are fewer $d\downarrow$ states to receive scattered spin \downarrow conduction electrons. A simple perturbation calculation [17] shows that the resistivity transfer (from ρ_{\downarrow} to ρ_{\uparrow}) depends on the direction of magnetization and is strongest for the magnetization parallel to the current; so we can write :

$$\begin{aligned} \rho_{\parallel\uparrow} &= \rho_{\perp\uparrow} + \gamma\rho_{\downarrow} \\ \rho_{\parallel\downarrow} &= \rho_{\perp\downarrow} - \gamma\rho_{\downarrow} \end{aligned} \quad (4)$$

where γ is a spin-orbit strength, assumed to be constant for all first series transition impurities. Substituting into expression (3), we get :

$$\frac{\rho_{\parallel} - \rho_{\perp}}{\rho_{\perp}} = \gamma \frac{\rho_{\downarrow}(\rho_{\downarrow} - \rho_{\uparrow})}{\rho_{\uparrow}\rho_{\downarrow} + \rho_{\uparrow\downarrow}(\rho_{\uparrow} + \rho_{\downarrow})} \quad (5)$$

and at low temperature ($\rho_{\uparrow\downarrow} \ll \rho_{0\uparrow}, \rho_{0\downarrow}$) :

$$\frac{\rho_{\parallel} - \rho_{\perp}}{\rho_{\perp}} = \gamma \left(\frac{\rho_{0\downarrow}}{\rho_{0\uparrow}} - 1 \right). \quad (6)$$

The experimental results for the resistivities at 4.2 °K of various alloys (Fig. 4) show that the rule (6) is well obeyed. When the temperature is increased, the variation of the anisotropy confirms well to the expression (5).

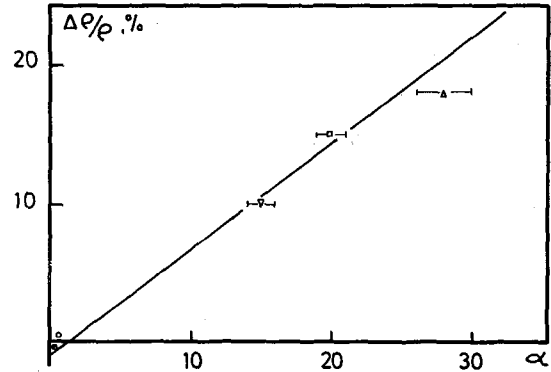


FIG. 4. — Experimental value of $\frac{\Delta\rho}{\rho} = \frac{\rho_{\parallel} - \rho_{\perp}}{\rho}$ at 4.2 °K in Ni alloys plotted as a function of the ratio $\alpha = \rho_{0\downarrow}/\rho_{0\uparrow}$. Straight line corresponds to $\Delta\rho/\rho = 0.0075(\alpha - 1)$. Note the negative experimental value for $\Delta\rho/\rho$ in NiCr ($\alpha_{\text{Cr}} - 1 = -0.6$).

The resistivity anisotropy has been shown to be strongly orientation dependant in monocrystals [19, 20]; a systematic study of this effect should give interesting information on the Fermi surface.

V. Thermoelectric power and thermal conductivity.

— The two current model has been shown to be essential in explaining the experimental values of T. E. P. and thermal conductivity in Ni alloys. A great deal of work has been done in this field by the group of Strasbourg and by Farrell and Greig.

In a two current model, the diffusion thermopower coefficient can be written

$$S = \frac{\sigma_{\uparrow} S_{\uparrow} + \sigma_{\downarrow} S_{\downarrow}}{\sigma_{\uparrow} + \sigma_{\downarrow}}$$

where σ_{\uparrow} and σ_{\downarrow} , S_{\uparrow} and S_{\downarrow} are the conductivities and T. E. P. coefficients for the spin \uparrow and spin \downarrow electrons.

Measurements on ternary alloys of Ni with Co and Cr [22] impurities led to precise values of the coefficients S_{\uparrow} and S_{\downarrow} associated with the scattering by Co or Cr impurities. The sign and the values of S_{\uparrow} for Cr have been used to estimate the width and the position of the virtual bound state in the spin \uparrow conduction band [22].

T. E. P. coefficients of the host metal for spin \downarrow electrons can be deduced from the variation of the T. E. P. with temperature in binary alloys [23, 24]. Phonon drag T. E. P. appears to be of opposite sign for the spin \uparrow and the spin \downarrow electrons [23].

The thermal conductivity is more difficult to explain completely as there is an important lattice contribution; however, self-consistent values can only be obtained if account is taken of the two current conduction [25].

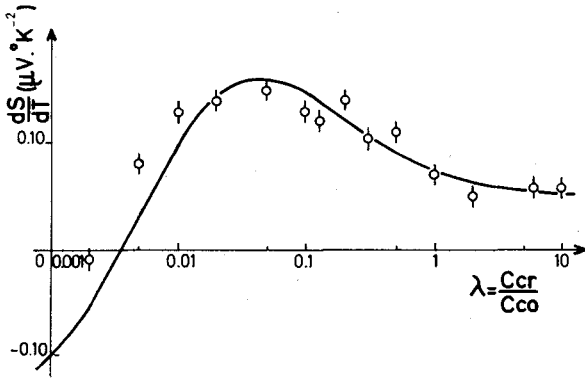


FIG. 5. — Experimental T. E. P. coefficients for NiCoCr alloys plotted as a function of relative concentration of Co and Cr. The full curve represents the variation calculated on a two current model with $S_{\uparrow} = -0.18 \mu\text{V}^{\circ}\text{K}^{-2}$, $S_{\downarrow} = 0$ for Co and $S_{\uparrow} = 0.25 \mu\text{V}^{\circ}\text{K}^{-2}$, $S_{\downarrow} = 0$ for Cr (after Cadeville et al. [22]).

VI. Hall effect (ordinary). — The low field ordinary Hall effect depends on the hole or electron character of the different carriers, weighted by their mobilities.

The experimental low temperature ordinary Hall effect appears to be very small for NiCo, NiFe [26] or NiMn [27] alloys for which the relaxation times are much longer on the spin \uparrow Fermi surface; this indicates [16] that for the spin \uparrow Fermi surface hole and electron character is more or less balanced.

Fairly detailed explanations [16] can also be given of the rapid variation of Hall coefficient with temperature and with impurity concentration.

VII. Magneto-resistance. — Polycrystal magneto-resistance results for dilute Ni alloys at low temperature have been shown [28, 29, 30] to fall on a set of different Kohler plots, depending of the major impurity present (the field parameter used for these plots has to be magnetic induction B , in agreement with De Haas Van-Alphen data [31, 32]). It is striking to observe (Fig. 6) that the magnetoresistance (both longitudinal and transverse) is much greater for samples where the impurity has a value of $\rho_{0\downarrow}/\rho_{0\uparrow}$ very

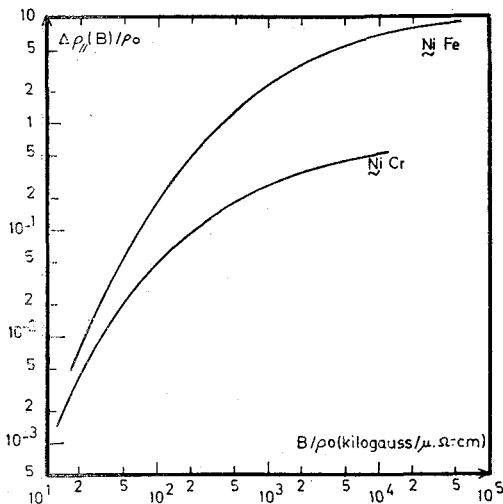


FIG. 6. — Kohler plots for the experimental longitudinal magnetoresistance in NiFe and NiCr alloys (Schwerer and Silcox [30]).

different from 1, that is where the current is mainly carried by electrons of one direction of spin. Thus for NiFe the ratio of saturation longitudinal magnetoresistance to zero B resistance is 10, an extremely high value, while that of NiCr is only about 1.5 [30].

Qualitatively, this can be understood from what is known about the band structure of Ni: the Fermi surfaces contain regions where spin-orbit effects hybridize spin \uparrow and spin \downarrow [33]; the magnetic field, by sweeping the electrons from the long relaxation time parts of the Fermi surface to regions where the spin-orbit coupling induces magnetic break-down between spin \uparrow and spin \downarrow surfaces, can produce a large increase of the resistivity. Experimental results for Ni alloys in the longitudinal high field limit correspond roughly to a resistivity enhancement by a factor $(\rho_{0\uparrow} + \rho_{0\downarrow})^2/4\rho_{0\uparrow}\rho_{0\downarrow}$ corresponding to complete spin mixing multiplied by a factor of about 1.5 associated with the usual relaxation time anisotropy.

A complicating effect is the reduction of the spontaneous anisotropy by the spin mixing induced by the field; this effect can explain some deviations from Kohler's rule at the beginning of the high field domain and also the decrease of spontaneous anisotropy in low concentration alloys [30] (where the internal induction $4\pi M$ is sufficient to produce spin mixing effects).

Another effect [3] is the suppression of spin waves by the magnetic field, which reduces the $\rho_{\uparrow\downarrow}$ term and therefore the variation of the resistivity with temperature for alloys where $\rho_{0\downarrow}/\rho_{0\uparrow}$ is very different from unity (NiCo, NiFe, NiMn).

Finally, at room temperature and above, there is a strong negative magneto-resistance due to a reduction by the field of spin disorder and possibly to band separation changes.

VIII. Extraordinary Hall effect. — This is the part of the Hall effect that depends on the magnetization of the sample, and is another manifestation of a spin-orbit interaction. Luttinger [34] proposed a mechanism which can be described as follows: an electron with magnetization \mathbf{m} and wave vector \mathbf{k} has a spin-orbit induced polarization $\mathbf{p} = \lambda\mathbf{k} \wedge \mathbf{m}$. In an electric field, the electron will have an energy term $\mathbf{E} \cdot \mathbf{p} = \lambda\mathbf{E} \cdot (\mathbf{k} \wedge \mathbf{m}) = \lambda\mathbf{k} \cdot (\mathbf{E} \wedge \mathbf{m})$.

The energy term is odd in k , and so will cause a displacement of the Fermi distribution; this is equivalent to a current in the direction perpendicular to \mathbf{E} and to \mathbf{m} , independent of the scattering processes (and so non-dissipative!). In the experimental set up, this is compensated by a voltage $V_{\perp} = \rho I_{\perp}$. Finally, the Hall coefficient is

$$R_s = \frac{V_{\perp}}{I_{\parallel} m} = \frac{\rho \lambda E m}{(E/\rho) m} = \lambda \rho^2.$$

This is found experimentally to be the dominant mechanism at room temperature. It has been pointed out by Kondorskii [35] that for a given spin direction, electrons and holes have opposite contributions to R_s . Roughly, R_s/ρ^2 can be taken to measure

$$\lambda[(n_{e\uparrow} - n_{h\uparrow}) - (n_{e\downarrow} - n_{h\downarrow})]. \quad (16).$$

At low temperatures, experiment shows [26, 27] that there exists an additional mechanism, which appears to be due to non-symmetric scattering processes

and gives an extraordinary Hall effect proportional to the resistivity (i. e. the concentration of impurities) and depending of the nature of impurities; in Ni the resulting R_s is positive for Cu, Fe [26] and Mn impurities but negative for Cr [27] impurities.

IX. High temperature effects. — We have so far discussed only the region $T \ll T_c$. Around and above T_c , comparison of resistivities for ferromagnetic and paramagnetic transition metals (Fig. 7) shows that spin disorder effects are important [36]. Results for $d\rho/dT$ in Ni show a Lambda peak at T_c ; a tail to this peak for $T > T_c$ shows that spin-spin correlations continue to exist above T_c . Fisher and Langer [37] suggest that (in Ni in any case) behavior is dominated by short range correlations—below T_c by zero range correlations, so magnetic resistivity depends on $\langle m_z \rangle^2$, and above T_c by the same short range correlations that dominate the enthalpy. Experimentally, above T_c , $d\rho/dT$ is proportional to C_v [38] and the magneto-resistance is proportional to the magnetocaloric effect [39]; both observations support the relation between magnetic disorder resistivity and enthalpy.

It would be interesting to fill in the gap between low temperatures, where spin disorder appears as the mixing term $\rho_{\uparrow\downarrow}$, and high temperatures where spin disorder is a direct resistivity.

X. Conclusion. — The transport properties of transition ferromagnetics display a wide range of interesting properties, specific to ferromagnetics,

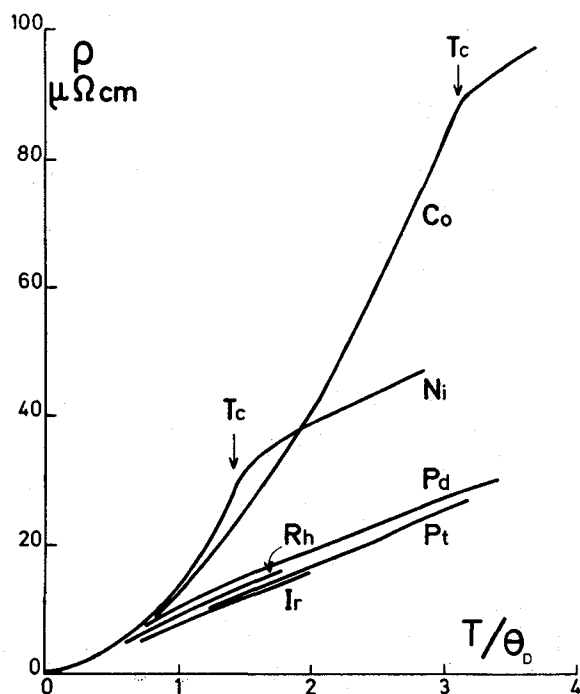


FIG. 7. — Resistivities of transition metals plotted against T/θ_D . This shows the excess resistivity due to spin disorder in Ni and Co.

which can on the whole be interpreted in a coherent and simple manner. Considerable further work, particularly on mono-crystals, is called for.

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