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## Transport properties of amorphous rare-earth alloys

R. Asomoza, I. A. Campbell and A. Fert

Laboratoire de Physique des Solides, Université Paris-Sud, Centre d'Orsay, 91405 Orsay, France

**Résumé.** — Nous discutons divers aspects des propriétés de transport des alliages amorphes de terres rares : contributions de l'ordre magnétique à la résistivité, magnétorésistance et son anisotropie, anomalies logarithmiques de résistivité à basse température, effet Hall extraordinaire.

**Abstract.** — We discuss several aspects of the transport properties of RE amorphous alloys : contributions from magnetic ordering to the resistivity, magnetoresistance and its anisotropy, low temperature resistivity anomalies, extraordinary Hall effect.

The transport properties of rare-earth (RE) amorphous alloys only begin to be studied. Some interesting effects have been observed : resistivity minima related to magnetic ordering and characteristic magnetoresistance, resistivity minima of non-magnetic origin in other systems, magnetic contributions to the Hall effect... Theoretical models have been proposed but not yet completely tested. We will present the topic in its present raw state.

### 1. Resistivity and magnetoresistivity of magnetic RE alloys. — 1.1 EXPERIMENTAL RESULTS ON Ni<sub>3</sub>RE AMORPHOUS ALLOYS [1, 2].

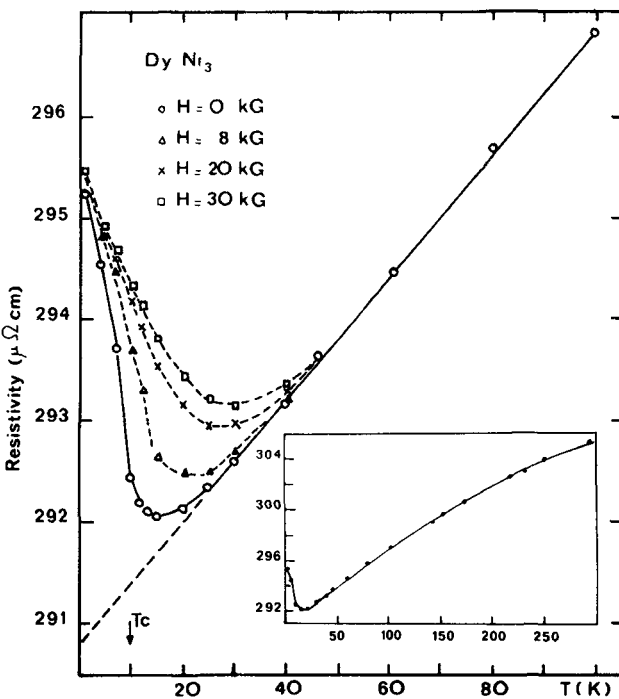


Fig. 1. — Resistivity of amorphous Ni<sub>3</sub>Dy versus temperature in several fields [1, 2].

resistivity of a Ni<sub>3</sub>Dy alloy as a function of the temperature. There is a minimum at  $T_M \approx 15$  K while the ordering temperature is about 10 K [2]. The change of resistivity between 1.2 K and the minimum is 1.1 %. Similar minima are observed in Ni<sub>3</sub>Ho and Ni<sub>3</sub>Er but the resistivity changes are smaller as the RE spin decreases suggesting an exchange scattering effect.

The magnetoresistance (figures 1 and 2) confirms the magnetic origin of the resistivity minimum. There is a positive magnetoresistance at low temperature with a peak in  $d\rho/dH$  near the ordering temperature. Well above this temperature the magnetoresistance becomes progressively weaker and takes approximately a dependence in  $\left(\frac{H}{T-\theta}\right)^2$  where  $\theta$  is close to the Curie-Weiss temperature of the paramagnetic susceptibility. The low temperature upturn of  $\rho$  and the positive magnetoresistance can be ascribed to the same mechanism : a positive contribution from

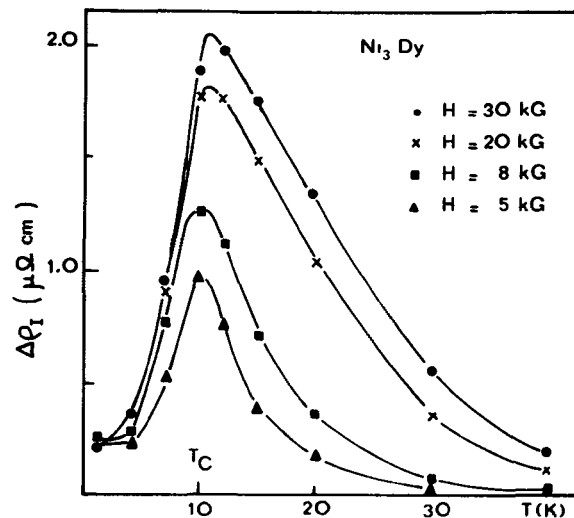


Fig. 2. — Magnetoresistance of amorphous Ni<sub>3</sub>Dy versus temperature at several fields [2] ( $\Delta\rho_1 = \frac{1}{3} \Delta\rho_{\parallel} + \frac{2}{3} \Delta\rho_{\perp}$ ).

magnetic ordering to  $\rho$ . The sign of the resistivity change with increasing magnetic order is however opposite to what is normally observed in crystalline alloys.

The  $\text{Ni}_3\text{Dy}$ ,  $\text{Ni}_3\text{Ho}$  and  $\text{Ni}_3\text{Er}$  behaviours are similar with only decreasing amplitudes as the RE spin decreases.  $\text{Ni}_3\text{Gd}$  shows a somewhat different behaviour on which we shall come back in section 1.3. Non-magnetic and magnetic Ni-Y alloys show resistivity minima which have a much smaller amplitude and are field independent (see section 2). Such non-magnetic effects may exist in  $\text{Ni}_3\text{Dy}$ ,  $\text{Ni}_3\text{Ho}$  or  $\text{Ni}_3\text{Er}$  but are certainly covered up by the magnetic contributions.

**1.2 RESISTIVITY FROM COHERENT EXCHANGE SCATTERING IN AMORPHOUS ALLOYS.** — The interference between waves scattered by the exchange interaction on neighbour magnetic ions contributes to the resistivity if there is some correlation between the spin directions. This so-called coherent-exchange scattering contribution has been calculated by de Gennes and Friedel [3] for crystalline ferromagnets. This calculation has been recently extended by Fert *et al.* [1, 2] to the case of amorphous alloys. Fert *et al.* [2] find the following expression of the resistivity due to exchange scattering :

$$\rho_m = \rho_M \left( \frac{J}{J+1} C_1 + C_1^2 m(2k_F) \right) \quad (1)$$

where

$$\rho_M = \frac{m^2 k_F \Gamma^2 J(J+1)}{4 \pi z e^2 \hbar^3} \quad (2)$$

$$m(q) = \frac{1}{NC_1^2 J(J+1)} \sum_{\mathbf{R}_0, \mathbf{R} \neq \mathbf{R}_0} e^{i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}_0)} \mathbf{J}_R \cdot \mathbf{J}_{R_0} \quad (3)$$

$C_1$  is the concentration of magnetic ions and  $\Gamma$  is the exchange constant.

The first term in the bracket of Eq. (1) gives the contribution from independent scattering by each ion (one-ion term). Actually, the factor  $J/(J+1)$  only holds for non-S ions in the low temperature range where the spin-flip scattering is frozen out by the anisotropy field (say below 50 K or 100 K). At high temperature this factor tends to unity.

The second term in the bracket of Eq. (1) gives the contribution from coherent exchange scattering. It is proportional to the number  $C_1^2$  of pairs and to the value at  $q = 2k_F$  of the spin correlation  $m(q)$ . As an amorphous structure is disordered except at short range the sum in  $m(q)$  is limited to neighbour pairs and thus  $m(q)$  depends on the *local magnetic order* (practically over 2 or 3 interatomic distances).

The calculation of the correlation function  $m(q)$  can be easily carried on when the range of the magnetic order is longer than the range of the structural order. For example, say that the sum in  $m(q)$  is limited to third neighbours and that the spin correlations are

the same between first, second and third neighbours :  $\langle \mathbf{J}_{R_0} \cdot \mathbf{J}_R \rangle = \mu_l^2 J^2$  where  $\mu_l$  is a parameter of local magnetic order. From eq. (1) and (3) one derives :

$$\rho_m = \frac{J}{J+1} \rho_M (C_1 + C_1^2 \{ a_{11}(2k_F) - 1 \} \mu_l^2) \quad (4)$$

where

$$a_{11}(q) = 1 + \frac{1}{NC_1^2} \sum_{\mathbf{R}_0, \mathbf{R} \neq \mathbf{R}_0} \exp(i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}_0)).$$

The meaning of Eq. (4) is very simple : the contribution from magnetic ordering ( $\mu_l \neq 0$ ) to  $\rho_m$  is positive when the interferences are constructive ( $a_{11}(2k_F) > 1$ ) and is negative when the interferences are destructive ( $a_{11}(2k_F) < 1$ ).

The calculation of  $m(q)$  is less simple when there are spin correlations of shorter range than the typical 2 or 3 interatomic distances (this occurs, for example, for antiferromagnetic interactions). It becomes then impossible to factorize  $\langle \mathbf{J}_{R_0} \cdot \mathbf{J}_R \rangle$  in Eq. (3) as above. In some cases a good approximation should be to limit the sum in  $m(q)$  to nearest neighbour pairs. For example constructive interferences between nearest neighbours (if  $\sin(2k_F r_{12}) > 0$  approximately) and antiferromagnetic correlations will now give a negative contribution to  $\rho_m$  (see section 1.4).

**1.3 RETURN TO THE EXPERIMENTAL RESULTS ON  $\text{Ni}_3\text{RE}$  ALLOYS.** — The results on  $\text{Ni}_3\text{Dy}$ ,  $\text{Ni}_3\text{Ho}$ ,  $\text{Ni}_3\text{Er}$  (figures 1 and 2) are in agreement with what is expected from Eq. (4) when  $a_{11}(2k_F) > 1$ . Asomoza *et al.* [1] have tried a quantitative fit and have shown that the resistivity changes corresponds to reasonable values of  $\rho_M$ . An interesting information is given by the weakness of the magnetoresistance well below  $T_c$  : it suggests that  $\mu_l$  approaches 1 and that, over few interatomic distances, there is not the asperomagnetic distribution of moments which is supposed to exist in a broader range [5] ; this could be due to correlations between the anisotropy axes of neighbour sites.

In the paramagnetic state, one expects

$$\mu_l \sim \langle J_z \rangle \sim H/(T - \theta)$$

in agreement with the experimental magnetoresistance  $\sim \left( \frac{H}{T - \theta} \right)^2$ . On the other hand a small resistivity upturn begins to appear above  $T_c$  and can be ascribed to the existence of short range order in the paramagnetic state. If this short range order corresponds to correlations between nearest neighbours Eq. (4) is no more valid and one should come back to Eq. (1) and (3) to account for the  $(T - \theta)^{-1}$  dependence of the upturn.

Bhattacharjee and Coqblin [6] have calculated the spin correlation function  $m(q)$  and the resulting resistivity in a model of two-spin cluster approximation with ferromagnetic interactions. They find the

resistivity upturn, the maximum of  $d\rho/dH$  at  $T_c$  and when they suppose parallel anisotropy axes on neighbours, they also find a fairly small magnetoresistance at  $T \ll T_c$  in agreement with the experimental data (figure 3).

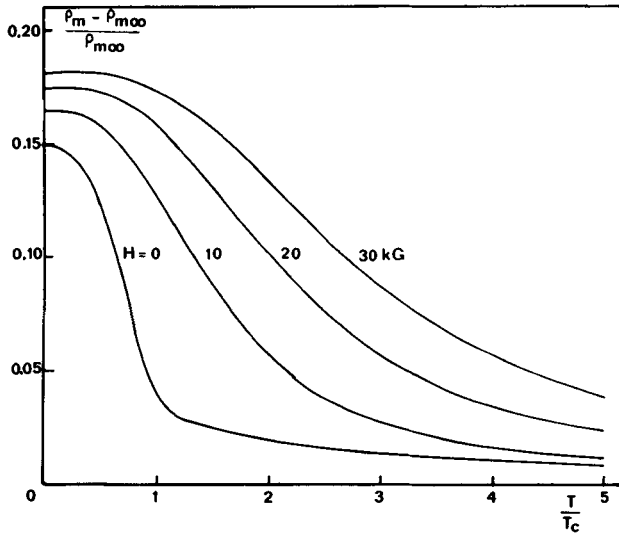


Fig. 3. — Magnetic resistivity versus  $T$  in several fields calculated by Bhattacharjee and Coqblin [6] in a two-spin cluster model with ferromagnetic interactions.

**1.4 RESISTIVITY AND MAGNETORESISTANCE OF AgRE ALLOYS AND SOME OTHERS.** — The resistivity of  $Ag_{1-x}RE_x$  amorphous alloys ( $0.2 < x < 0.5$ ) shows an upturn at low temperature [7] (figure 4). The upturn appears to be again of magnetic origin as it occurs at about the ordering temperature. Measurements

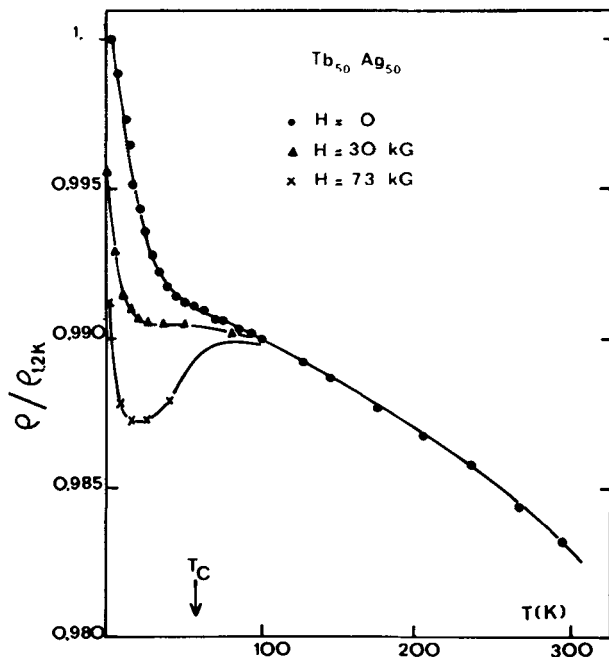


Fig. 4. — Resistivity of amorphous  $Ag_{50}Tb_{50}$  versus temperature in several fields [7].

have been made on series of alloys in order to check the approximately parallel variation of the temperatures of the magnetic ordering and the resistivity upturn when the concentration or the factor  $(g - 1)J$  changes. There is also a strong magnetoresistance at low temperature but, in contrast to what occurs in  $Ni_3Re$ , this magnetoresistance is negative (figure 5).

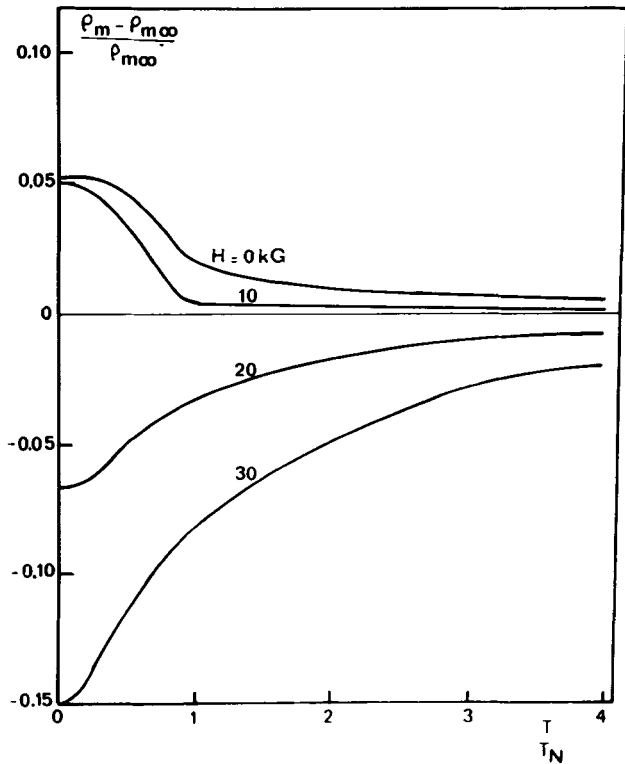


Fig. 5. — Same caption as figure 3 but antiferromagnetic interactions.

This behaviour is coherent with predominant antiferromagnetic interactions and destructive interferences. An applied field would then lower  $\rho$  by aligning the spins while the antiferromagnetic correlations of the ordered state would raise  $\rho$ . The antiferromagnetic character of the interactions in the AgRE alloys has been already proposed by Bhattacharjee and Coqblin [8] to explain the magnetic properties. They have now applied their two-spin cluster model with antiferromagnetic couplings to calculate the correlation function and the resistivity. The agreement is reasonably good (figure 5). In our opinion however this interpretation is not yet sure. Other interpretations can be also proposed : for example dominant ferromagnetic interactions (in agreement with the positive Curie-Weiss temperature) but largest interference effects corresponding to the distance of antiferromagnetic pairs. It would be certainly interesting to calculate the resistivity in other models proposed for the magnetic structure.

The resistivity of  $Au_{20}(La_{1-x}Gd_x)_{80}$  alloys has been also studied [9]. For  $x < 0.4$  the results are roughly

similar to those in Ag-RE (upturn of  $\rho$  at low temperature and negative magnetoresistance) and could be interpreted in the same way by coherent exchange scattering. For the  $x = 1$  alloy in which the order turns out to be ferromagnetic, there is, on the contrary, a decrease of  $\rho$  below  $T_c$ . This change of behaviour is of course coherent with a change from antiferromagnetic to ferromagnetic correlations. On the other hand Poon *et al.* [9] have proposed another interpretation which attributes the resistivity upturn to Kondo-like scattering by magnetic clusters. For the moment it is difficult to conclude.

There are also data on the resistivity of  $Al_2RE$  alloys [10]. In  $Al_2Gd$  and  $Al_2Dy$  there is a large excess of resistivity at low temperature with respect to  $Al_2La$  and  $Al_2Y$ . Is this excess of magnetic origin? It is difficult to conclude without magnetoresistance data. However note that the resistivity excess extends well above the temperature  $T_\theta$  of the susceptibility cusp. This is in agreement with the idea [11] of  $T_0$  being a freezing temperature of ferromagnetic clusters formed at higher temperature.

Resistivity data also exist for alloys such as Co-RE and Fe-RE. Generally  $d\rho/dT$  is negative. As these alloys have a very high  $T_c$  the magnetic contribution to the resistivity must be displayed on a wide temperature range and it is certainly difficult to separate in  $d\rho/dT$  the magnetic contribution from the normal contribution due to the thermal variation of the structure factor (Ziman theory). We believe that this normal contribution should be predominant in the majority of cases.

**1.5 ANISOTROPY OF MAGNETORESISTANCE.** — This effect has been studied in detail for dilute RE impurities in noble metal hosts [12]. The large anisotropy in these crystalline alloys is due to the electronic quadrupole of the RE  $f$  shell;  $\rho_{\parallel} - \rho_{\perp}$  is about zero for Gd and changes sign half-way through the heavy RE series.

In  $Ni_3RE$  amorphous alloys [2] the anisotropy of magnetoresistance is again extremely small for  $Ni_3Gd$ , exists for  $Ni_3Dy$ ,  $Ni_3Ho$  and  $Ni_3Er$  (figure 6) but is only about 10% of the isotropic part of the magnetoresistance (studied in 1a, see figure 2). The anisotropy is positive for  $Ni_3Dy$ ,  $Ni_3Ho$  and negative for  $Ni_3Er$  demonstrating the quadrupolar origin.

The weakness of the anisotropy in  $Ni_3RE$  is related to the random crystal field. The quadrupolar polarization  $\langle J_z^2 \rangle - J(J+1)/3$  is zero for a truly hemispherical distribution of moments (asperomagnetism) and becomes different from zero if the exchange or applied field closes the angle of the distribution. On figure 6 the applied field is responsible of the high field slopes and the exchange field gives the non-zero intercept of the high field slopes to  $H = 0$  at low temperature. The value at 1.2 K of the spontaneous anisotropy (intercept at  $H = 0$ ) in  $Ni_3Dy$  corresponds to a distribution of moments in a cone

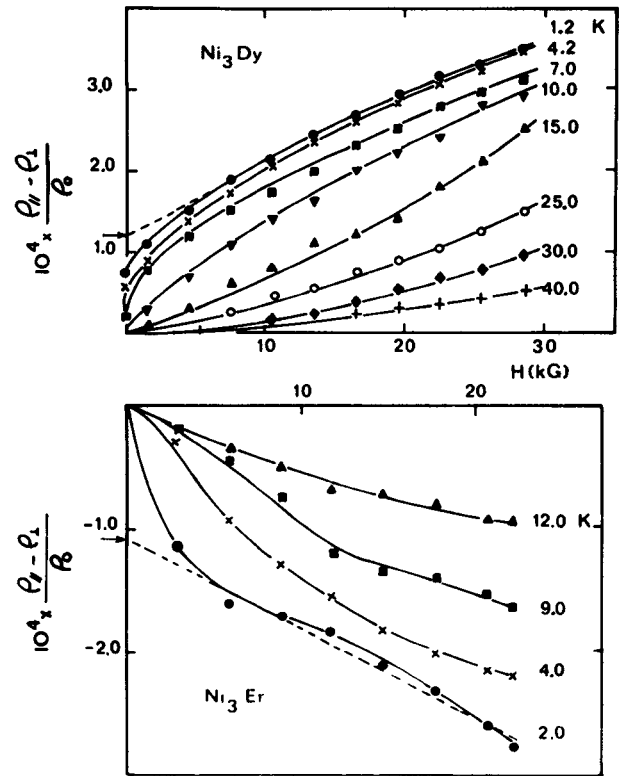


Fig. 6. — Resistivity anisotropy of  $DyNi_3$  and  $ErNi_3$  [2].

of angle  $89^\circ$  (an asperomagnetic structure corresponds to  $90^\circ$ ). Some data on the resistivity anisotropy also exist for RE-transition alloys [13] and Ag-RE alloys [7].

**2. Low temperature resistivity anomalies of non-magnetic origin.** — **2.1 GENERAL.** — Resistivity variations of the form  $-\ln T$  are observed at low temperature in many amorphous alloys such as CoP, NiP or Metglas alloys [14]. Recently such variations have been also observed in magnetic  $Ni_{95.6}Y_{4.4}$  and  $Ni_{93}Y_7$  alloys [14]. This is one of the few examples of logarithmic anomalies in amorphous alloys containing only metallic elements. More recently similar

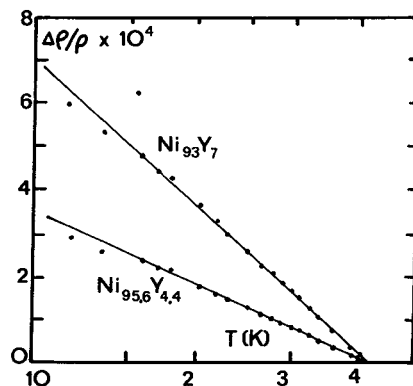


Fig. 7. — Relative resistivity of  $Ni_{95.6}Y_{4.4}$  and  $Ni_{93}Y_7$  versus  $\ln T$  [15].

anomalies have been also observed in non-magnetic  $\text{Ni}_3\text{Y}$  [2] and  $\text{AgLu}$  [7] alloys.

Figure 7 shows for  $\text{NiY}$  alloys [14] the good agreement with a variation in  $-\ln T$  with some deviations below 1.5 K. Such deviations and finally a saturation below 1 K are classically observed in CoP or Metglass alloys. This variation is not changed by a magnetic field which led to ascribe it to scattering by two level systems of structural origin [16]. In contrast the resistivity variation in alloys containing magnetic RE (preceding section) was very field dependent and also larger by an order of magnitude.

**2.2 THEORETICAL PROBLEMS.** — The amorphous materials show several characteristic anomalies in their specific heat and acoustic properties at low temperature. These anomalies have been explained by the existence of two level systems associated to the tunnelling between atomic configurations of nearly equal energy. Cochrane *et al.* [16] have ascribed the logarithmic resistivity to the electron scattering by these two-levels systems. They write the scattering potential

$$H_1 = \sum_{\mathbf{k}\mathbf{k}'\alpha\alpha'=\pm 1} V_{\mathbf{k}\mathbf{k}'}^{\alpha\alpha'} a_{\mathbf{k}\alpha}^\dagger a_{\mathbf{k}'\alpha'} b_\alpha^\dagger b_{\alpha'} \quad (5)$$

where  $b_\pm^\dagger$  ( $b_\pm$ ) are creation (annihilation) operators of tunnelling states + or - and  $a_{\mathbf{k}\pm}^\dagger$  ( $a_{\mathbf{k}\pm}$ ) are creation (annihilation) operators of conduction states  $|\mathbf{k}_\pm\rangle$ . An important assumption is to take conduction waves  $|\mathbf{k}_\pm\rangle$  already distorted by the scattering potential and partly orthogonal ( $|\mathbf{k}_\pm\rangle$  being the distorted wave when the two level system is in the state +). Thus the first scattering stage is phenomenologically treated. The second stage is the scattering of the distorted waves by  $H_1$  and is similar to the Kondo scattering in magnetic alloys. One obtains a resistivity term of third order with respect to  $H_1$

$$\rho \sim -\ln(kT^2 + \Delta^2) \quad (6)$$

where  $\Delta$  is the difference of energy between the tunnelling states. If  $kT \gg \Delta$ ,  $\rho \sim -\ln(T)$ .

Kondo [17] has treated also this problem. His perturbation treatment of the scattering (first and second stages of the calculation of Cochrane *et al.* together) gives :

$$\rho = C \left( \ln \frac{T}{D} \right)^2 \quad (7)$$

where  $C$  is of the fourth order with respect to the scattering potential. Thus the two theoretical treatments give two different results :  $\ln T$  and  $(\ln T)^2$ . Moreover the experimental results turn out to be in better agreement with the result of the semi-phenomenological model of Cochrane *et al.* rather than with the apparently more rigorous model of Kondo. In fact resistivity minima seem to occur always in alloys with transition elements and, owing to the strong

potential of transition atoms, the semi-phenomenological model might be more appropriate than the perturbation treatment.

**3. Hall effect.** — The Hall resistivity of magnetic materials is expressed as  $\rho_H = R_o B + R_s 4 \pi M$  where  $R_o B$  is the ordinary Hall resistivity and  $R_s 4 \pi M$  is the extraordinary Hall resistivity. In the amorphous magnetic alloys the extraordinary Hall effect is largely predominant, which can be understood by the following argument. As the extraordinary Hall effect results from orbital exchange or spin-orbit coupling through asymmetries of the scattering it is an increasing function of the number of scatterers and then of the resistivity (the contributions from skew scattering and side-jump are roughly proportional to  $\rho$  and  $\rho^2$ ). The extraordinary Hall effect is thus very strong in amorphous alloys having a high resistivity (<sup>1</sup>) while the ordinary Hall constant remains near  $(\text{Ne})^{-1}$ . This gives  $\rho_H$  practically proportional to the magnetization  $M$  (the Hall effect can be used to record hysteresis loops, for example).

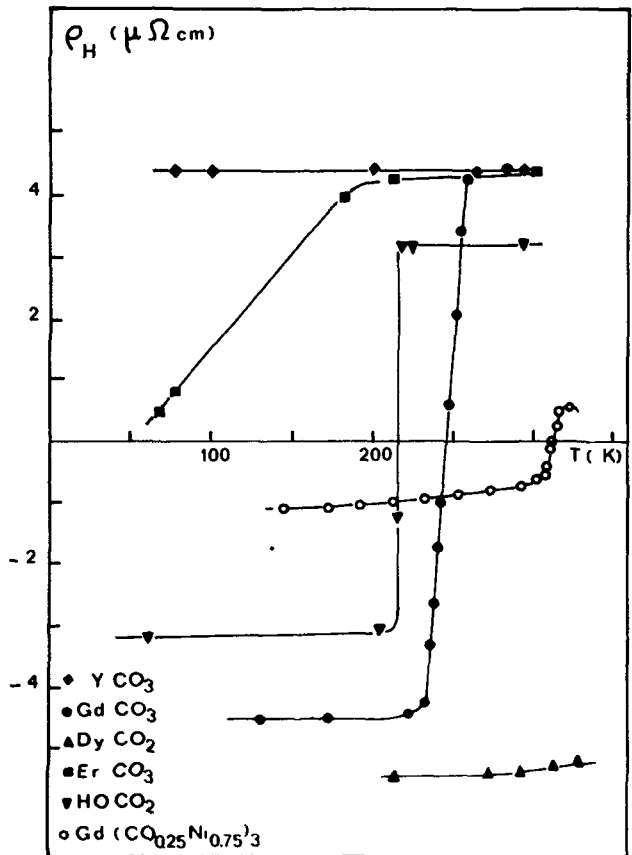


Fig. 8. — Hall resistivity of several amorphous alloys versus temperature [19].

(<sup>1</sup>) Some non-magnetic liquid or amorphous alloys (liquid La or amorphous LaGa alloys for example) show a positive Hall effect suggesting an extraordinary contribution from the susceptibility of the conduction electrons. Such an effect in non-magnetic alloys can be expected in transition and some heavy metals.

The extraordinary Hall effect has been mainly studied in RE-transition alloys [13], [18], [19] and some data also exist for RE-noble metal alloys [13]. An interesting result is the change of sign of the Hall effect at the compensation temperature of ferromagnetic RE-Co alloys (figure 8). This change of sign can be understood by assuming independent contributions from the Co and the RE. Mc Guire *et al.* [13] write

$$\varphi = c_{\text{RE}} \varphi_{\text{RE}} m_z^{\text{RE}}/M_{\text{RE}} + c_{\text{Co}} \varphi_{\text{Co}} m_z^{\text{Co}}/M_{\text{Co}} \quad (8)$$

where  $\varphi = \rho_{\text{H}}/\rho$  is the Hall angle,  $\varphi_{\text{RE}}$  and  $\varphi_{\text{Co}}$  are the Hall angles of hypothetical pure and fully ordered RE and Co,  $c_{\text{RE}}$  and  $c_{\text{Co}}$  are the concentrations,  $m_z$  is the magnetization along the field and  $M$  is the saturation magnetization. Below  $T_{\text{comp}}$   $m_z^{\text{RE}} > 0$ ,  $m_z^{\text{Co}} < 0$  while above  $T_{\text{comp}}$   $m_z^{\text{RE}} < 0$ ,  $m_z^{\text{Co}} > 0$ , what explains the change of sign. Mc Guire *et al.* [13] have tried to determine constant values of  $\varphi_i$  for some elements and to use equations similar to Eq. (8) to account for the Hall angles of hypothetical pure and fully ordered agreement is reasonably good. We however believe

that such an analysis is very approximate. Firstly Eq. (8) holds only for the contribution from skew scattering while other forms of equation are expected if the mechanism is side-jump scattering by the orbital exchange with localized moments or side jump scattering of polarized electrons by spin-orbit interactions. Secondly the change of the conduction band with the concentration is ignored in Eq. (8). As a matter of fact the extension of Eq. (8) with constant coefficient  $\varphi_i$  to large series is difficult and, for example, the Hall resistivity of  $\text{Co}_3\text{RE}$  or  $\text{Co}_2\text{Dy}$  in ref. [19] are different from those predicted by Eq. (8) with the parameters  $\varphi_i$  of ref. [13]. A better understanding of the microscopic mechanisms is needed to make progress in the field.

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