

# HALL EFFECT IN FERROMAGNETIC Tb<sub>75</sub>-Gd<sub>25</sub>. ELECTRON SCATTERING AND CRITICAL BEHAVIOUR (\*)

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**ABSTRACT** — The Hall effect resistivity ( $\rho_H$ ) of a ferromagnetic alloy has been investigated in a Tb<sub>75</sub>-Gd<sub>25</sub> sample (hcp structure;  $T_c = 244$  K), in the temperature range 77-300 K and with applied magnetic fields up to  $7.96 \times 10^5$  A.m<sup>-1</sup> (10 KOe). A reversal in the sign of  $\rho_H$  has been observed at  $T^* = 175$  K, suggesting the existence of competing electron scattering mechanisms. We analysed such effect in terms of the skew and side-jump contributions.

The magnitude of the Hall resistivity over the whole ferromagnetic phase investigated could be well described in terms of the Hall resistivities of pure Tb and Gd.

The high accuracy of our data enabled the observation of spin-fluctuation effects near the Curie point of Tb<sub>75</sub>-Gd<sub>25</sub>. In particular we obtained the quantity  $d\rho_H/dT$ , which exhibits the critical features associated with a second-order magnetic transition. A preliminary mean-field analysis is made in the paramagnetic phase.

## 1 — INTRODUCTION

The Hall resistivity  $\rho_H = E_H/j$  ( $E_H$  = transverse electric field;  $j$  = longitudinal current density) results from the transverse motion of the electrons under an applied magnetic field  $H_a$ , which produces a magnetic induction  $B$  inside the sample. In a normal metal, the Hall resistivity is entirely due to the Lorentz force on the electrons and thus simply related to  $B$ :

$$\rho_H^o = R_o \cdot B \quad (1)$$

$R_o$  is the so called Hall constant.

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In a magnetic metal, a new contribution appears due to the finite magnetization  $\mathbf{M}$  in the cooperative phase, and one then writes:

$$\rho_H = \rho_H^o + \rho_H^s = R_o \cdot B + \mu_o \cdot R_s \cdot M \quad (2)$$

$\rho_H^s$  is the "extraordinary" Hall resistivity and  $R_s$  is the extraordinary Hall constant ( $\mu_o =$  vacuum magnetic permeability). Usually in heavy rare earths we have  $R_s \gg R_o$  (one or two orders of magnitude). This new term is caused by the existence of several asymmetric scattering processes ( $\alpha$ ) of the electrons by the lattice spins  $\mathbf{S}_i$ , through representative interaction Hamiltonians  $H_i^{(\alpha)}$  [1, 2]. An electron with a wave vector  $\mathbf{k}$  has then a different probability of being deflected to one or the other side of the plane ( $\mathbf{k}, \mathbf{S}_i$ ).

One possible mechanism in the localized spin systems of concern here (heavy rare earths) is the spin-orbit coupling  $\lambda_1 \mathbf{l} \cdot \mathbf{S}_i$ , where  $\mathbf{l}$  is the electron orbital angular momentum and  $\lambda_1$  is an interaction field [1, 2]. As shown by Kondo [1], another important asymmetric term exists, of the form  $\lambda_2 (\mathbf{l} \cdot \mathbf{S}_i) (\mathbf{s} \cdot \mathbf{S}_i)$ , where  $\mathbf{s}$  is the electron spin and  $\lambda_2$  an appropriate interaction field. Both terms originate preferential angular deflections for the electrons ( $\mathbf{k} \rightarrow \mathbf{k}'$ ), and so two distinct contributions for the Hall resistivity (skew scattering).

Besides such angular deflections, there is a purely quantum mechanical effect associated with the electron interaction (either of type  $\lambda_1$  or  $\lambda_2$ ): the corresponding wave packet effectively suffers a small transverse displacement  $\delta \mathbf{r}$  with respect to its initial direction, the so called side-jump effect [3, 4]. This originates an additional contribution to the transverse current, and thus to the Hall effect.

If we calculate the Hall resistivity within the first Born approximation for the  $\lambda_1$  and  $\lambda_2$ -skew scattering, one always finds  $\rho_H = 0$ . One has to go into the second Born approximation to obtain a finite contribution to the skew scattering Hall resistivity. The side jump contribution to  $\rho_H$  can be calculated by recalling that, for each collision, an electron suffers a transverse displacement  $\delta r_{\perp}$ , to which we can associate an effective transverse velocity  $\delta r_{\perp} / \tau$ , where  $\tau$  is the average time between consecutive collisions.

The calculations show that [5, 6]: (i) for the  $\lambda_1$ -skew scattering (S. S.) and the side jump effect (S. J.) the Hall resistivity keeps the same sign over the whole cooperative phase (ii) for the  $\lambda_2$ -term the Hall resistivity changes sign once, as T increases from 0 up to the magnetic critical point  $T_c$ . All these contributions may be simultaneously present, and this can bring considerable difficulties to separate them out of the measured Hall resistivity. In magnetic systems like Gd, GdAl<sub>2</sub>, PrAl<sub>2</sub>, NdAl<sub>2</sub> [7, 8, 9] the Hall resistivity exhibits the same sign over the whole temperature range, and this indicates the predominance of the  $\lambda_1$  contributions. On the other hand, in elements like Tb or Dy, the Hall resistivity changes from negative values near  $T_c$  to positive ones at lower temperatures [10, 11, 12].

In this context it is very useful to study magnetic alloys formed by elements which, when isolated, display one of these two types of behaviour. In the work reported here we study a ferromagnetic Tb<sub>75</sub>-Gd<sub>25</sub> alloy, formed by Tb, which exhibits a reversal in the sign of  $\rho_H$  (when alone), with Gd, for which  $\rho_H$  exhibits an intrinsic negative sign over the whole ferromagnetic phase. Quantitatively, it is also of interest to correlate the magnitude of  $\rho_H$  in the alloy at each temperature with the corresponding values for the elements Tb or Gd.

Besides the problem of the sign and magnitude of  $\rho_H$  in the alloy, we also gave particular emphasis to the investigation of the critical behaviour of the Hall resistivity in the vicinity of the Curie point  $T_c$ . This is best achieved through the measurement of the temperature derivative  $d\rho_H/dT$  in the transition region, which requires fairly high resolution in the  $\rho_H(T)$  measurements. To the best of our knowledge, this brings a novel contribution to the available experimental studies on the Hall effect (see also ref. [7]).

## 2 — RESULTS AND DISCUSSION

High accuracy data on  $\rho_H(T)$  were obtained with a lock' in technique [7], using a.c. currents of about 0.5 A through the sample, and applied magnetic fields up to  $7.96 \times 10^5$  A.m<sup>-1</sup> (10 KOe).

The ferromagnetic sample was a thin slab of  $Tb_{75} - Gd_{25}$  with dimensions  $0.22 \times 2.65 \times 9.1 \text{ mm}^3$ , and the measurements were performed in the temperature range 77-300 K, at several values of the applied field ( $H_a$ ). This field was set perpendicular to the slab plane. Inversion of the applied field was made at each experimental point to extract the odd part of the transverse voltage, i.e. the Hall voltage.

As an illustration we show in Fig. 1 the  $\rho_H(T)$  curves with  $H_a = 3.88 \times 10^5$  and  $7.72 \times 10^5 \text{ A.m}^{-1}$ . Similar measurements have been performed at  $H_a = 0.80 \times 10^5$  and  $1.59 \times 10^5 \text{ A.m}^{-1}$ .

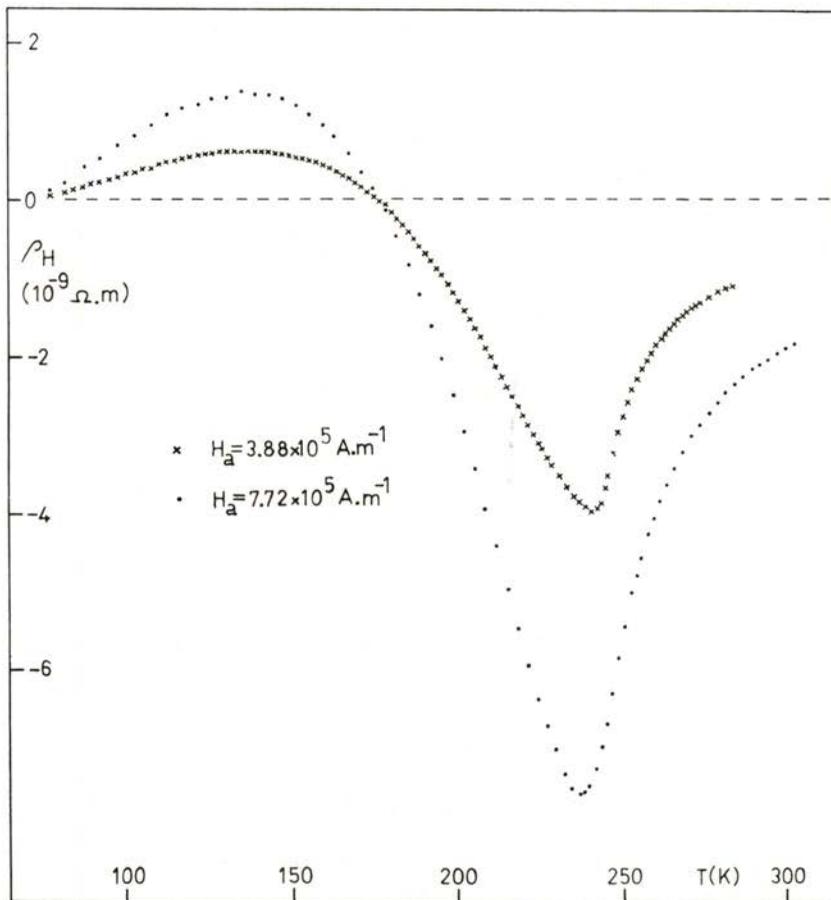


Fig. 1 — Hall resistivity of a polycrystal of  $Tb_{75} - Gd_{25}$ , as a function of temperature ( $T$ ) for two different values of the applied magnetic field ( $H_a$ ).

The Hall resistivity vanishes and changes sign at  $T^* = 175$  K, becoming positive below this temperature, with a broad maximum around 135 K. The observation that  $\rho_H(T^*) = 0$ , irrespective of the magnetic field, is consistent with the usual separation of the Hall resistivity as a sum of a normal ( $\rho_H^0$ ) and an extraordinary contribution ( $\rho_H^s$ ). In fact, under our experimental conditions,  $H_a$  is too small to produce magnetic saturation in the sample at  $T^*$  [13], and so the average magnetization is then given by [7]:

$$M = H_a/D \quad (3)$$

where  $D$  is the demagnetization factor of the sample ( $D \simeq 1$  in our geometry). Then we get (from eq. 2):

$$\rho_H(T) = \mu_0[R_o(T) + R_s(T)] \cdot (H_a/D) \simeq \mu_0[R_o(T) + R_s(T)] H_a \quad (4)$$

It becomes clear that  $T^*$  is just the temperature at which the quantity inside the brackets vanishes, i.e.

$$R_s(T^*) = -R_o(T^*) \quad (5)$$

The rapid decrease of  $\rho_H(T)$  in the paramagnetic phase is due to the corresponding decrease of the induced magnetization, by thermal disorder. More precisely, the induced magnetization above  $T_c$  is given by

$$M(T, H_a) = \chi(T) [1 + D\chi(T)]^{-1} \cdot H_a \quad (6)$$

where  $\chi(T)$  is the magnetic susceptibility in the paramagnetic phase (defined by  $\chi = M/H_i$ , where  $H_i$  is the internal magnetic field,  $H_i = H_a + (1 - D) \cdot M$ ). In this equation the denominator takes account of the effect of the demagnetizing field. Then the quantity  $\rho_H$  measured at constant  $H_a$  is approximately given by (from eq. 2):

$$\rho_H \simeq \mu_0 R_o H_a + \mu_0 R_s \cdot \chi(T) [1 + D\chi(T)]^{-1} \cdot H_a \quad (7)$$

At  $T$  sufficiently above  $T_c$ , i.e. when the short range spin-spin correlations become negligible, both  $R_o$  and  $R_s$  are independent of temperature, and so the decrease of  $\rho_H$  is entirely imposed by the decrease of  $\chi(T)$ . The magnitude of  $\rho_H$  at constant  $T$  should then increase linearly with  $H_a$ , as supported by the experimental results (see Fig. 1). The same linearity is observed at sufficiently

low temperatures, when the applied field becomes insufficient to saturate the sample ( $M \approx H_a$ , see eq. 3; and eq. 4 for  $\rho_H \propto H_a$ ).

Near the Curie point,  $R_o$ ,  $R_s$  and  $M$  are strongly dependent on the applied field, due to the field effect on the spin-spin correlations, and thus  $\rho_H$  ceases to be a linear function of  $H_a$ . This is clearly shown in Fig. 2, with  $\rho_H$ -data corresponding to  $H_a = 0.8 \times 10^5$ ,  $1.59 \times 10^5$  and  $7.72 \times 10^5$  A.m $^{-1}$ .

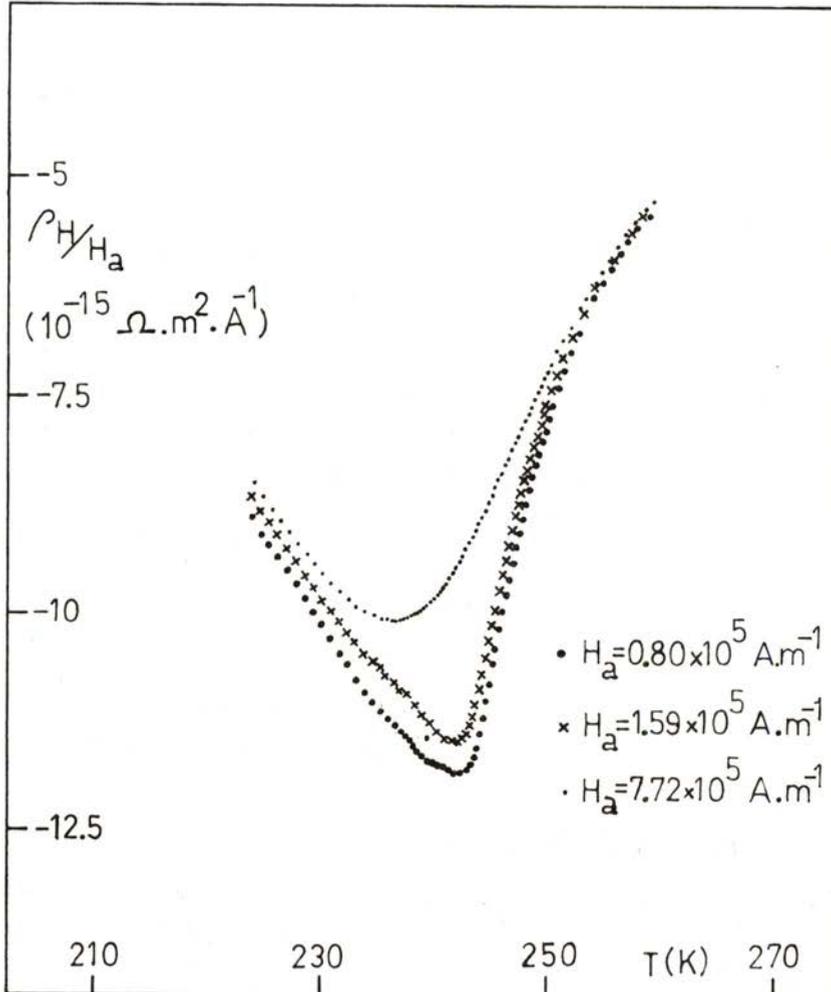


Fig. 2 — Near the Curie point the Hall resistivity ceases to be proportional to the applied magnetic field.

The general shape of  $\rho_H(T)$  in polycrystalline Tb<sub>75</sub>-Gd<sub>25</sub> is remarkably close to the curve obtained from an interpolation of the  $\rho_H(T)$  curves for polycrystalline Gd and Tb [14], weighted by the appropriate atomic percentages and with  $T$  scaled by the corresponding  $T_c$  values (Fig. 3), i.e.

$$\rho_H(T/T_c)_{\text{alloy}} = 0.75 \rho_H(T/T_c)_{\text{Tb}} + 0.25 \rho_H(T/T_c)_{\text{Gd}} \quad (8)$$

One can now add some comments on the possible role of the different skew and side jump contributions. The  $\lambda_2$ -contributions are expected to be fairly small, since they result directly from a second order effect, i.e. from the polarization of the conduction electrons by the lattice spins. For the case of pure Gd, for which a complete separation has been achieved recently [9], the small magnitude of the  $\lambda_2$ -contributions has indeed been confirmed. In a first approximation we neglect these contributions in Tb<sub>75</sub>-Gd<sub>25</sub>. Since none of the  $\lambda_1$ -contributions changes sign in the ferromagnetic phase, when alone, the experimental verification of a reversal in the sign of  $\rho_H$  for Tb<sub>75</sub>-Gd<sub>25</sub> clearly indicates that both  $\lambda_1$ -contributions (skew and side jump) are important in this alloy and have opposite signs. A more elaborate analysis of these results is in progress, with an extension of the theory of the Hall effect to the case of magnetic alloys.

With regard to the critical behaviour, Fig. 4 displays the temperature derivative  $d\rho_H/dT$  as a function of temperature, the derivative being obtained by "local" (3-point) numerical differentiation of the  $\rho_H(T)$ -data. For convenience the data have been normalized by a factor  $\rho_H(T_c)$ . The existence of "critical features" in the Hall effect is evidenced by the very sharp dip in  $d\rho_H/dT$  at  $T = 244.0$  K. This value is in good agreement with the Curie temperature previously obtained with Tb<sub>75</sub>-Gd<sub>25</sub> single crystals, using magnetization ( $T_c = 245 \pm 2$  K) [13] and electrical resistivity studies ( $T_c = 244.8 \pm 0.5$  K) [15]. Also, the minimum in  $1/\rho_H(T_c) \cdot (d\rho_H/dT)$  at  $T_c$  gets less sharp as  $H_a$  increases, as expected from the reduction of the spin-spin fluctuations caused by the increase in  $H_a$ .

As mentioned above, the role of these fluctuations diminishes markedly as  $T$  increases in the paramagnetic phase (in heavy rare earths, they can be neglected for  $\varepsilon \gtrsim 10^{-1}$ , with  $\varepsilon = (T - T_c)/T_c$ ;

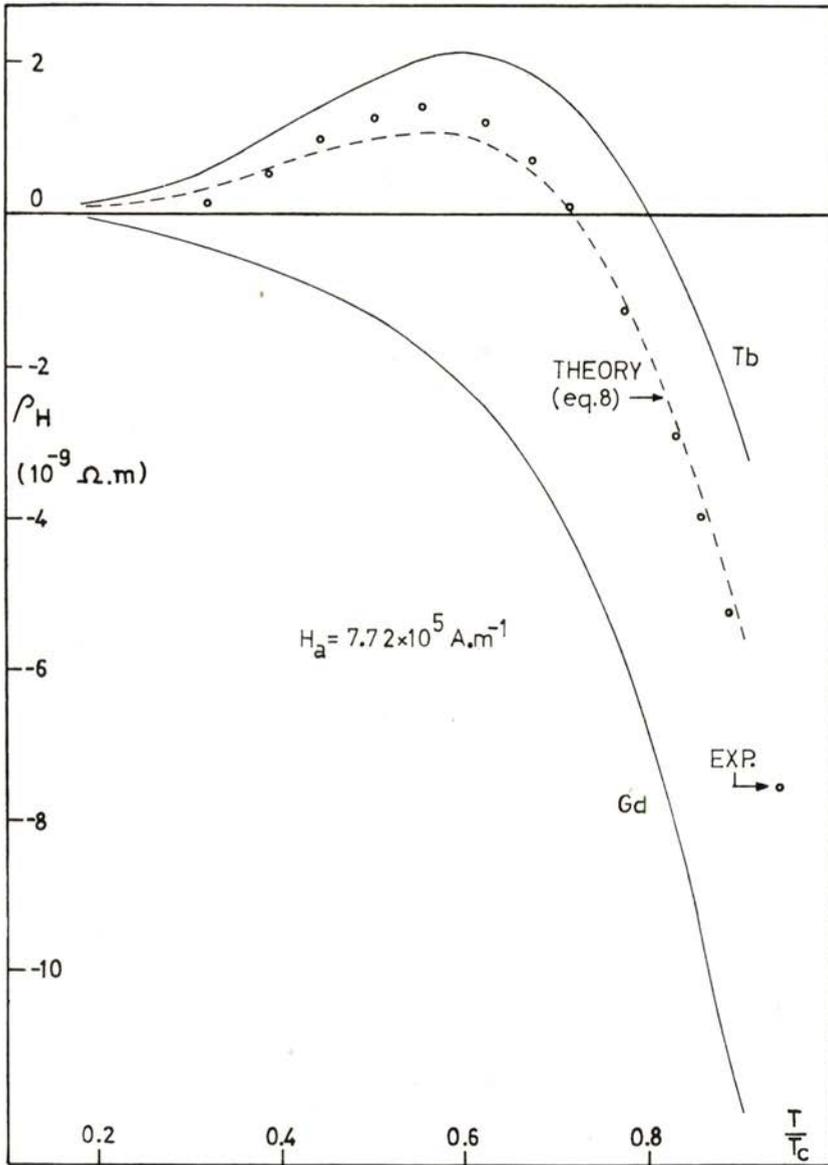


Fig. 3 — Comparison between the experimental Hall resistivity ( $\rho_H$ ) of  $Tb_{75} - Gd_{25}$  and the theoretical prediction based on the Hall resistivities of polycrystalline Tb and Gd (eq. 8).

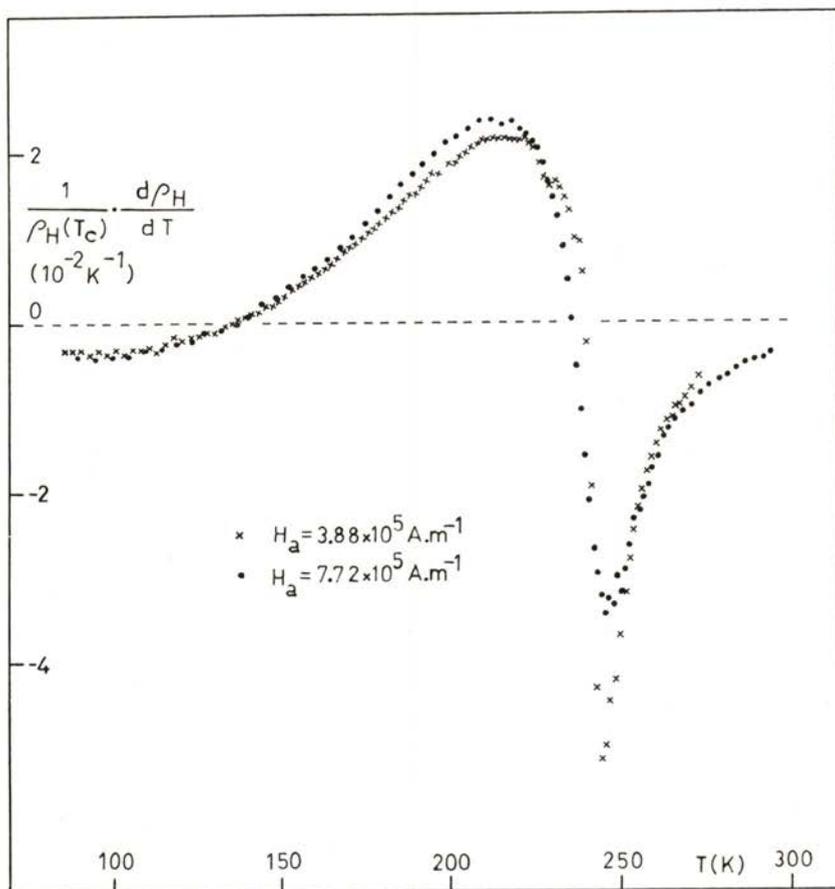


Fig. 4 — Temperature dependence of the thermal derivative,  $d\rho_H/dT$ , of the Hall resistivity, normalized by  $\rho_H(T_c)$ ;  $T_c = 244$  K.

see e.g. ref. [15]), and so  $R_o$  and  $R_s$  are expected to become constant. Then eq. (4) predicts

$$\rho_H \simeq R_o \mu_o H_a + R_s \mu_o \chi H_a \tag{9}$$

Assuming, for  $T$  sufficiently above  $T_c$ , that  $\chi$  obeys a Curie-Weiss law [13],

$$\chi = C / (T - T_c^*) \tag{10}$$

where  $T_c^*$  is the paramagnetic Curie-Weiss temperature and  $C$  the Curie-Weiss constant, the expression (9) takes the form:

$$\rho_H(T) = A + B / (T - T_c^*) \quad (11)$$

where  $A$  and  $B$  are constants.

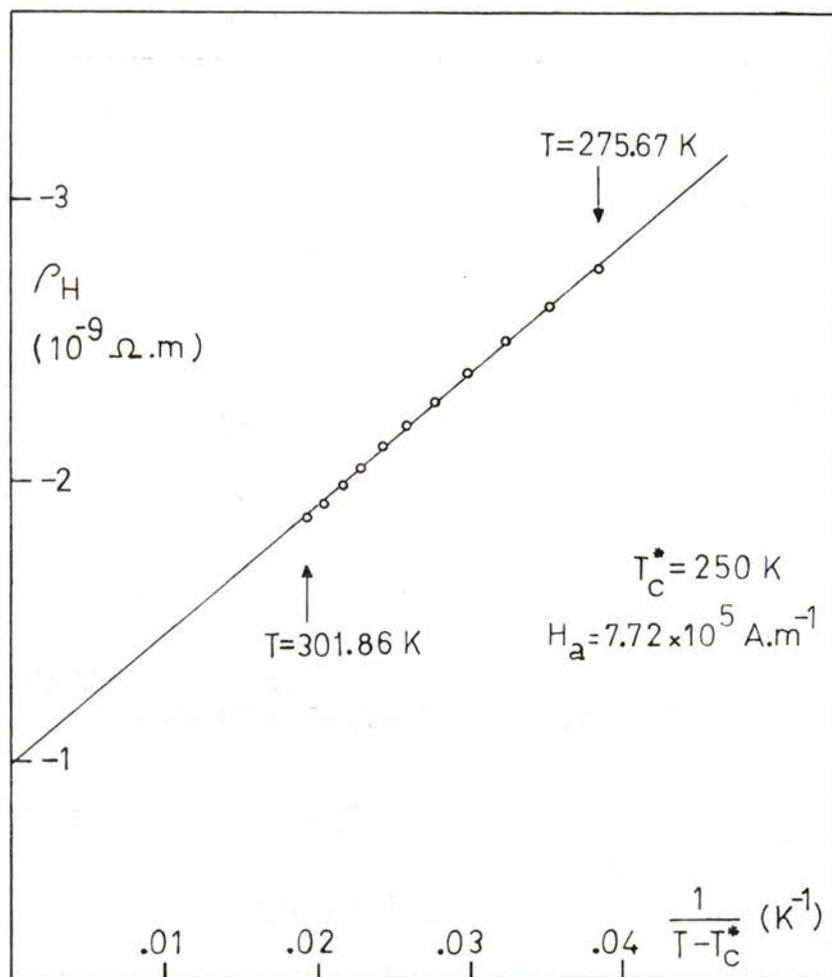


Fig. 5 — Hall resistivity of  $Tb_{7.5} - Gd_{2.5}$  versus  $(T - T_c^*)^{-1}$ , where  $T_c^*$  is the paramagnetic Curie temperature ( $T_c^* = 250 \text{ K}$ ); from the linear plot we obtain the constants  $A$  and  $B$  of eq. (11).

In order to check this prediction we performed a numerical analysis of our  $\rho_H$ -data above 275 K ( $\varepsilon > 1.27 \times 10^{-1}$ ), with  $T_c^*$  considered as an adjustable parameter. For a chosen  $T_c^*$  value, a least squares fitting to expression (11) gave the values of A and B with the corresponding relative errors and the correlation parameter ( $\sigma$ ), indicative of the quality of the fitting. The correlation  $\sigma$  was found to be maximum for  $T_c^* = 250.0 \pm 0.25$  K ( $\sigma = 0.9985$ ) and the errors in A and B were found minima at that temperature ( $A = (1.00 \pm 0.02) \cdot 10^{-9} \Omega m$ ,  $B = (45.97 \pm 0.86) \cdot 10^{-9} \Omega m K$ ). The good quality of the corresponding fit can be seen in Fig. 5.

Further data analysis is in progress for temperatures very close to the Curie point, in order to extract relevant information on the critical behaviour of the Hall effect. Also, the experimental investigation of the relation between the critical behaviour of  $\rho_H(T)$  and that of the magnetic susceptibility  $\chi(T)$  is being contemplated.

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Note added in proof: A recent analysis [9] has shown that the Smit asymmetric scattering mechanism (J. Smit, *Physica*, **21**, 877 (1955)) gives a fairly large contribution to  $\rho_H$  in Gd; it is likely that such mechanism may also be operative in Gd-Tb alloys.