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Hall Effect of Yb Intermediate Valent Intermetallic Compounds

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Hall effect measurements between 300 K and 4.2 K for the intermediate valent intermetallics YbCuAl, $YbCu_2Si_2$, $YbInAu_2$, YbPd, $YbAl_3$ and $YbAl_2$ are presented and discussed in detail. Correlations found between the Hall coefficient, the electrical resistivity and the magnetic susceptibility for some of these compounds suggest an anomalous contribution due to skew scattering.

1. Introduction

The Hall effect of intermetallic Yb and Ce compounds showing valence instabilities has been studied intensively during the last three years [1, 2]. As it is well known these compounds show anomalies in the transport properties which include large and temperature dependent electrical resistivities and thermopowers. While today most of the relevant experimental features concerning the thermopower and the electrical resistivity are already familiar, the phenomenology for the Hall effect still brings some new unexpected features worth of being studied.

In this paper we give a detailed discussion of Hall effect measurements for the well known Yb intermetallic intermediate valent compounds (IV) YbCuAl, $YbCu_2Si_2$, $YbInAu_2$, YbPd, $YbAl_3$ and $YbAl_2$. These compounds, which cover a wide range of values for the Yb valency from Yb^{3+} to Yb^{2+}, give the possibility for a systematic study of Hall anomalies and to build up correlations with other physical properties.

The experimental details and the sample preparation is given in Sects. 2.1 and 2.2 respectively.

A description of the experimental results considering correlations between the Hall coefficient, the electrical resistivity and the magnetic susceptibility is given in Sect. 3.

The discussion in Sect. 4 is divided into two parts. The first Sect. 4.1 deals with general phenomenological features, and the second Sect. 4.2 includes a discussion in terms of the theory of the Hall effect in IV compounds recently proposed by Ramakrishnan et al. [3, 4].

All the measured Yb compounds with the exception of the relatively new YbPd are already classical intermediate valent compounds; most of their physical properties have already been studied. Relevant work on these materials may be found in the proceedings of the intermediate valence conferences [5–8] and in review articles [9, 10]. The valences of the measured Yb compounds from L_{III} absortion may be found in [11, 12].

2. Experimental

2.1. Hall Measurements

The Hall Voltage V_H was measured in the conventional four leads ac technique with a Lock-In amplifier (Ithaco Dynatrac 391A) and a preamplifier (Ithaco model 165), providing a resolution $\delta V_H \sim 2$ nVolts. The off-set voltage V_0 due to the misalignment of the contacts was minimized ($V_0 < 10.V_H$) by careful contacting of the samples and finally suppressed with the zero-suppress of the Lock-In.

The ac current was supplied by an oscillator (Krohn-Hite model 4100), the operating frequency being $\nu = 117$ Hz, with a maximum current intensity of 65 mA.

The samples were cut into a rectangular form with a low speed diamond saw. The length to width ratio

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was not smaller than 3 in order to avoid that a possible reduction of the measured transverse voltage due to short circuiting by the current leads (soft welded along the ends of the samples) exceed a few per cent of the neal Hall voltage as shown in [13].

The samples were ground down to a thickness d between 200 and 300 microns using boron carbide. d was measured with a micrometer ($\delta d = 50$ microns). Copper leads (diameter $\varnothing = 50$ microns) were soft welded to the samples with In as indicated in [1, 14]. The Hall voltage contacts were made as small as possible (≤ 100 microns) to avoid short circuiting and local distortions of the sample thickness. The spacing between Hall contacts was almost equal to the width of the sample (typical 2.7–3 mm) so that no corrections were necessary.

The absolute sign of the Hall coefficient R_H was determined by means of a dc measurement with the usual sign convention [15]. The measuring temperature range from 1.5 K up to 300 K was covered with two cryostats: one for the temperature interval between 1.5 K and 60 K [16], with a maximum available field $H_{max} = 50$ KOe) and a second one for the temperature interval between 77 and 300 K [14] with $H_{max} = 8$ KOe. V_H was measured at fixed temperatures by increasing the field in regular steps in both field directions, the Hall voltage then being given by the average.

In the temperature range between 1.5 K and 60 K the magnetic field was measured with a longitudinal Hall sensor (Lake Shore model LHGA-321), between 77 K and 300 K with a gaussmeter (Bell 640), and the temperature with a carbon glass resistor (Lake Shore CGR) and a silicon diode (Lake Shore DT 500-P), respectively. Details about the experimental set-up may be found in [14]. The errors in measuring T and H are estimated to be 1%, and the error in determining the low field Hall coefficient R_H is 7%. The reproducibility was better than 2% for measurements on the same sample.

The electrical resistivity in ac was measured for the same samples as described in [14, 17] with an estimated error of 5%.

2.2. Sample Preparation

The polycrystalline samples were prepared by melting together stoichiometric amounts of the elements in an induction furnace on a water cooled copper crucible in a purified Ar atmosphere of about 1300 torr. For compounds $YbCu_2Si_2$ (tetragonal $ThCr_2Si_2$), $YbInAu_2$ (β Wolfram A_2) and $YbAl_2$ (cubic $MgCu_2$) an extra amount of about 8% of Yb was added in order to compensate losses due to preferential evaporation of this element.

Debye Scherrer powder diffraction patterns of the samples showed no foreign phases with the exception of $YbAl_2$ were faint lines belonging to the cubic Cu_3Au (010) and (111) reflexes of $YbAl_3$ were identified.

The $YbInAu_2$ sample was annealed in a sealed quarz tube at 800 C for 2–4 days, thus lowering the residual resistivity value from 10.3 to 2.3 $\mu\Omega$ cm. The samples for $YbAl_2$ and $YbCu_2Si_2$ were not annealed to prevent evaporation of Yb and cracking, respectively. The samples for $YbAl_3$ (Cu_3Au), YbPd (CsCl) and YbCuAl (Fe_2P) were prepared by D. Müller [17], B. Politt [11] and R. Pott [18] respectively.

3. Results

The electrical resistivity ρ measured for the same samples used for the Hall measurements is shown in Fig. 1. $\rho(T)$ shows the well known non linear temperature dependence with a strong saturation for YbCuAl and $YbCu_2Si_2$. The saturation of the electrical resistivity for $YbInAu_2$ and $YbAl_3$ is more pronounced at higher temperatures [17]. A general compilation of resistivities for IV compounds may be found in [19], studies of the saturation effect for some of these materials are given in [19] and [20].

Figure 2 shows the low field Hall coefficient R_H for YbCuAl and $YbCu_2Si_2$ which have a negative and a positive sign respectively. Disregarding the different signs they show some similar qualitative features: an increase of R_H in magnitude with decreasing temperature in the temperature region where $\rho(T)$ shows strong saturation and a maximum or a plateau at intermediate temperatures (clearly seen in Figs. 2 and 4 in Ref. 2). R_H for $YbCu_2Si_2$ decreases steeply below 20 K, in the same temperature interval where ρ settles into its residual resistivity ρ_0 (see also Fig. 1 in [2]). Compared to other stable valent RCu_2Si_2 compounds with R=Gd, Tb and Y (see Fig. 2 in [2]) R_H for $YbCu_2Si_2$ has a much stronger temperature dependence and is larger. The same is also valid for the R_H of YbCuAl with respect to the R_H for its nonmagnetic reference compound LuCuAl; R_H(LuCuAl) 0.32×10^{-3} cm^3/C between 300 and 77 K [14]. LuCuAl and YbCuAl have different signs in R_H, however, a suitable extrapolation of R_H (YbCuAl) for high temperatures shows that at high temperatures both compounds are positive (see sect. 4). Figure 3 shows R_H for $YbInAu_2$, $YbAl_3$, $YbAl_2$ and $LuAl_3$. The Hall coefficient for $YbInAu_2$ changes sign from negative to positive at about 160 K, while $LuInAu_2$ has a negative weakly temperature dependent R_H of about -0.11×10^{-3} cm^3/C. Disregarding the sign change, R_H for $YbInAu_2$ and $YbCu_2Si_2$ are qualitatively similar as shown in Fig. 3

of [2]. $R_H(YbInAu_2)$ shows a plateau below 70 K and also a steep decrease (below 25 K) in the temperature interval where ρ settles into ρ_0. The plateaus for R_H for $YbCu_2Si_2$ and $YbInAu_2$ and the peak in YbCuAl occur in the same temperature intervals where $\rho(T)$ shows approximately a linear temperature dependence (inflexion point), see also Figs. 1,3,4 in [2]. This temperature interval tends to be narrower for the compounds approaching Yb^{3+} ion.

For $YbAl_3$ the anomaly in R_H develops also in the direction of more positive values like in $YbCu_2Si_2$ and $YbInAu_2$, however $R_H(T)$ does not show the same curvature as it is almost linear in T between 300 and 90 K (Figs. 7 and 3). A similar feature with the former compounds is the steep decrease of R_H in the same temperature interval where ρ settles into ρ_0, i.e. below 50 K (Fig. 7).

R_H for $YbAl_2$ shows a weaker temperature dependence than the other Yb compounds in Fig. 3, showing a relative change of 34% in R_H between 300 and 77 K. The Hall coefficient at room temperature $R_H = 0.29 \times 10^{-3}$ cm^3/C is relatively close to the value known for $LaAl_2$ at room temperature $R_H = 0.43 \times 10^{-3}$ cm^3/C [21] and is also consistent with the values of R_0 extrapolated from $R_H(T)$ for $T \to \infty$ for the magnetic RAl_2: $R_0 = 0.24$, 0.36, 0.33 and 0.22×10^{-3} cm^3/C For R=Tb, Gd, Pr and Nd respectively [21]. This features suggest that there are no significant anomalies in R_H for $YbAl_2$ in the temperature range of the measurement. The electrical resistivity, shows also no significant deviations from the linear temperature dependence expected from phonon scattering (Fig. 1). The anomalous temperature dependence for $\rho(YbAl_2)$ develops above 300 K [17, 22].

The features mentioned above show correlations between ρ and R_H. One is the steep decrease in magnitude of R_H in the temperature interval where ρ settles into ρ_0. The other is that the absence of a high temperature anomalous saturation in the electrical resistivity corresponds to a weak temperature dependence for R_H. It would be tempting to generalize this and say that a strong saturation in ρ goes together with a strong temperature dependence of R_H like in YbCuAl and $YbCu_2Si_2$, but there is at least one exception: the magnetically ordered IV compound YbPd. This compound, which was studied intensively recently [11, 23], shows more complex physical properties than the more classical IV described previously. We mention the following properties [11]:

i) It orders magnetically (probably antiferro) at 0.5 K. Magnetic order is an unusual property in Yb IV compounds, particularly surprising in YbPd considering its high valency of 2.8. It is the only Yb compound reported here showing magnetic ordering.

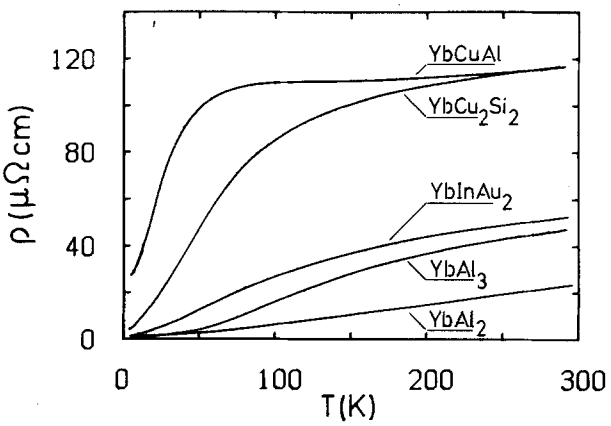

Fig. 1. Temperature dependence of the electrical resistivity ρ for YbCuAl, $YbCu_2Si_2$, $YbInAu_2$, $YbAl_3$ (D. Müller [17]) and $YbAl_2$

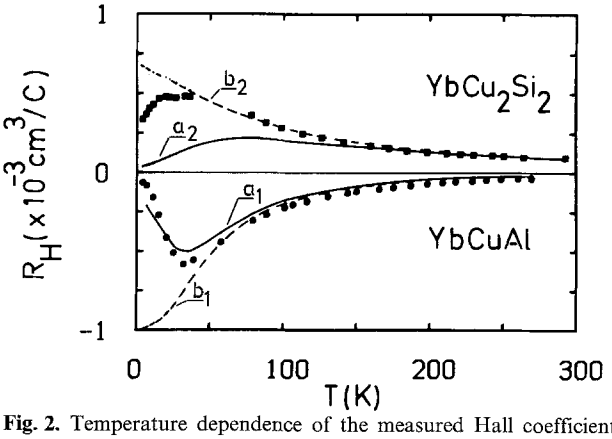

Fig. 2. Temperature dependence of the measured Hall coefficient R_H for $YbCu_2Si_2$ ■ and YbCuAl ●. Curves a_1, a_2 show R_H calculated using $\rho_b = \rho(T)$, and curves b_1, b_2 using ρ_b=constant in the expression (3) of Sect. 4.2

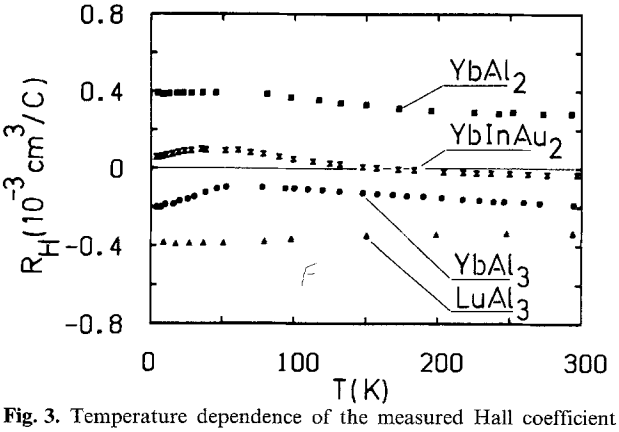

Fig. 3. Temperature dependence of the measured Hall coefficient R_H for $YbAl_2$ ■, $YbInAu_2$ ▼, $YbAl_3$ ● and the reference compound $LuAl_3$ ▲

ii) The electrical resistivity shows a very strong saturation above 140 K at a large value of about 140 μΩ cm.

R_H for YbPd (Fig. 4) is negative and weakly temperature dependent. The strongest temperature de-

pendence is observed below 18 K, where a monotonous increase in magnitude of R_H with decreasing T is observed. This feature is not shown by the compounds described previously; their Hall coefficients tend to approach at low temperatures the high T values (see Figs. 2 and 3). In YbPd the weak temperature dependence for R_H between 140 and 300 K is remarkable, it does not correlate as above with the strong saturation of $\rho(T)$. There are however some qualitative similarities between R_H for YbCuAl above 10 K and R_H for YbPd above 100 K and their resistivities above these temperatures. The striking difference is that the maximal relative change for R_H for YbPd between 100 and 300 K amounts only 25% of the room temperature value.

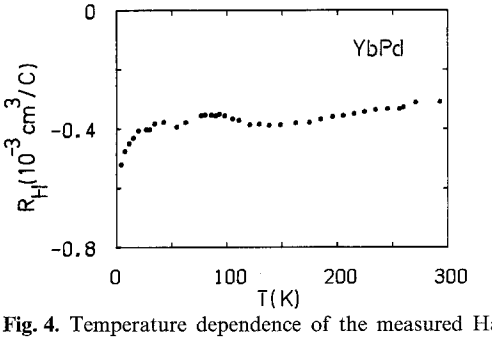

Fig. 4. Temperature dependence of the measured Hall coefficient R_H for YbPd

It is possible to understand qualitatively this discrepancy by correlating the Hall coefficient with the magnetic susceptibility. A comparison between these two quantities is reasonable in magnetic materials because the Hall anomalies may be due to an extraordinary Hall effect.

The magnetic susceptibility X for YbCuAl, $YbCu_2Si_2$, $YbInAu_2$ and $YbAl_3$ taken from the literature, is shown in Fig. 5.

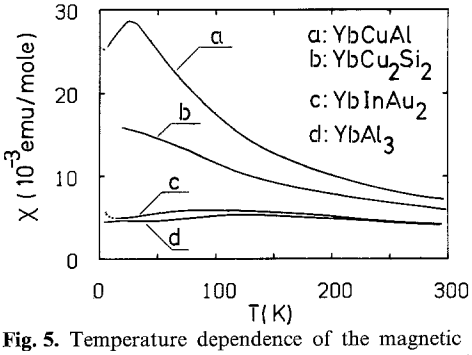

Fig. 5. Temperature dependence of the magnetic susceptibility X for YbCuAl [24], $YbCu_2Si_2$ [25], $YbInAu_2$ [26] and $YbAl_3$ [27]

Comparing Fig. 5 with Figs. 2 and 3 we notice that the strong temperature dependence of R_H for YbCuAl and $YbCu_2Si_2$ correlates with their relatively large and strongly temperature dependent X, and also that $YbInAu_2$ and $YbAl_3$ show a weaker temperature dependence and smaller absolute values for $X(T)$ and $R_H(T)$ than the former.

Consistent with these observations $YbAl_2$ shows almost no temperature dependence in $R_H(T)$ and $X(T)$ (0.42×10^{-3} emu/mole [27]).

To summarize, the Yb Intermetallics with weakly temperature dependent susceptibilities have also weakly temperature dependent Hall coefficients.

The magnetic susceptiblity of YbPd between 100 and 300 K lies in between that of $YbCu_2Si_2$ and $YbInAu_2$ being also smaller and more weakly temperature dependent than the susceptibility of YbCuAl. This explains in part the quantitative discrepancy pointed out before.

The description of the phenomenology given above shows that relevant features like the temperature dependence and the magnitude of R_H are directly related to $\rho(T)$ and $X(T)$, supporting an interpretation of the Hall anomalies in terms of an extraordinary Hall effect. A quantitative discussion is given in Sect. 4.2.

The Hall resistivities $\rho_H(H)$ of the measured Yb IV compounds show a linear field dependence up to $H_{max} = 50$ KOe. Slight deviations from linearity were only observed in $YbCu_2Si_2$ and YbCuAl below 8 K amounting 3% and 10% respectively at 4.2 K and 50 KOe, and in YbPd below 18 K, with 20% at 7 K and 50 KOe. YbPd shows the strongest field dependence in ρ_H.

A last important point in this description of results is the sign of the R_H anomaly. Comparing Figs. 2, 3, and 4 it is clear that all the measured Yb compounds develope anomalies in the direction of positive R_H with the exception of YbCuAl and YbPd which show negative anomalies. One difference between these two compounds and the others is that in the temperature region where $\rho(T)$ saturates, $d\rho/dT$ is smaller than the phonon slope of the corresponding stable valent references [24, 11]. However a clear-cut correlation to predict the sign of the anomaly in R_H was not found.

4. Discussion

4.1. General Features

The relationships between R_H, ρ and X suggest the possibility of explaining the observed anomalies by means of an extraordinary Hall effect.

The low field Hall coefficient R_H for paramagnetic materials is given by the empirical form [28]:

$$R_H = R_0 + R_s X. \tag{1}$$

Where R_0 is the "normal Hall coefficient" due to the deflection of the conduction electrons by the Lor-

entz force. R_s is the "anomalous Hall coefficient" which depends on the concentration of the magnetic scatterers c and on the parameters of some mechanism for asymmetric scattering related to spin orbit-coupling [29]. In the presence of a magnetic field the conduction electrons are then scattered preferentially to the right or to the left of the magnetic ions, rendering macroscopically an extra contribution to R_0. If R_s is proportional to c the extraordinary Hall effect is due to skew scattering, if it is proportional to c^2 it is due to a side-jump mechanism [29]. There is evidence for both effects in rare earths elements and alloys [30, 31].

Assuming the validity of (1) it is possible to derive some general conclusions without considering any particular model.

As already reported in [2] R_H for YbCuAl, $YbCu_2Si_2$ and $YbInAu_2$ may be fitted with:

$$R_H = R_0 + A/(T-\theta_H) \qquad (2)$$

A, θ_H (equivalent to $-\theta$ in [2]) and R_0 are constants (see Fig. 6).

The form (2) cannot be applied to all measured systems as R_H for $YbAl_2$, $YbAl_3$ and YbPd do not follow (2). Figure 6 shows however the following relevant features.

i) The inverse of $\Delta R_H = |R_H - R_0|$ shows a monotonic temperature dependence above some characteristic temperature T_{ch} (see Table 1 in [2]). Above T_{ch} the compounds with smaller susceptibility (higher valency) have a smaller Hall anomaly ΔR_H.

ii) The expression (2) fits the $R_H(T)$ for YbCuAl above 80 K with a positive R_0 (Table 1, third column) indicating the possibility of a sign change at high temperatures. A positive normal Hall coefficient R_0 for YbCuAl is also consistent with the positive R_H of LuCuAl (Table 1).

iii) A main feature is that $\Delta R_H(T)$ is not proportional to $X(T)$ with a constant factor, because in the temperature range where (2) is valid $X(T)$ follows a Curie-Weiss law with a large negative Curie temperature ($\theta_c = -34$ K and -90 K for YbCuAl and $YbCu_2Si_2$ respectively), whereas the intercepts obtained from Fig. 6 give positive temperatures. Then if (1) also holds R_s must be temperature dependent.

The physical properties of IV and Kondo systems are usually described considering a characteristic energy or temperature (see for example [32, 33, 34]) separating a low from a high temperature regime. T_{ch} may be defined through a maximum of some Physical property, the linewidth of the neutron scattering quasielastic line [35], or as a fit parameter taken from an ansatz function (T_{sf} in [32]). In a formal way a T_{ch} is given by the energy associated with some microscopic interaction proposed in a model [34, 36]. Estimates of a characteristic temperature based on the susceptibility, the T_{sf} of Wohlleben and Sales [32], are given in Table 1.

Table 1.

Compound	T_{sf}	R_0 [a]	R_0 [b]	R_H (reference)
YbCuAl	150	0.07	0.046	0.20 (LuCuAl)
$YbCu_2Si_2$	80	0.01	0.013	0.12 (YCu_2Si_2)
$YbInAu_2$	124	−0.07	−0.071	−0.11 ($LuInAu_2$)
$YbAl_3$	160	–	−0.270	−0.33 ($LuAl_3$)
Units	K	$\times 10^{-3}$ cm^3/C		

[a] After (2) in Sect. 4.1
[b] After (3) in Sect. 4.2

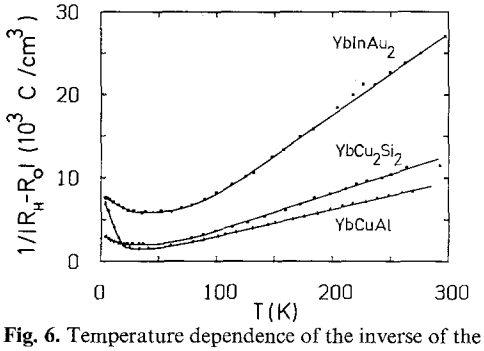

Fig. 6. Temperature dependence of the inverse of the absolute value of the Hall anomaly $R_H - R_0$ for $YbInAu_2$ (+), $YbCu_2Si_2$ (+) and YbCuAl (−) with R_0 values given in the second column of Table 1

To investigate the dependence of R_s on the resistivity a mobility ratio defined as $\Delta R_H/\rho$ is useful. For the compounds listed in Table 1 the mobility as function of the temperature shows a monotonic increase with decreasing temperatures, the only exception being YbCuAl which shows a maximum at 20 K. The mobility ratios for YbCuAl, $YbCu_2Si_2$, $YbInAu_2$ and $YbAl_3$ have the same order of magnitude (10 cm^2/Volt sec), and it is possible to scale them using some adecuate characteristic temperature [37]. A general difficulty in finding a relationship between R_s and ρ is to separate the contributions not giving asymmetric scattering from the total measured resistivity. At high temperatures ($T > T_{ch}$), however, ρ is mainly due to the anomalous scattering of the IV Yb ions [17] which usually predominates against the other scattering contributions which give the residual and the phonon resistivities ρ_{ph}. For example ρ_{ph} amounts only 10% of ρ in $YbCu_2Si_2$ [17]. This anomalous scattering is probably asymmetric as suggested by the orders of magnitude for ρ necessary to fit the Hall anomalies as shown in Sect. 4.2.

At low temperature the situation might not be the same. It seems, for example, that ΔR_H does not depend on the residual resistivity produced by static defects: two different pieces of the same $YbInAu_2$ sample annealed at different temperatures having $\rho_0 = 2.3$ μΩ cm and 7.6 μΩ cm respectively showed al-

most the same R_H. In a compound like YbCuAl with a high residual resistivity it is not clear which amount is intrinsic to the Yb ions and which due to static defects. By alloying non magnetic impurities into Yb compounds we expect a drastic change in R_H only when the valence state of the Yb ions is changed. A possible effect of the phonon scattering in the low temperature ΔR_H is discussed in Sect. 4.2.

We find a main difference when comparing the extraordinary Hall effect of stable with that of unstable ions. In paramagnetic alloys or magnetically ordered compounds with stable magnetic moments it is possible to describe ΔR_H in the paramagnetic range with an expression equivalent to (1) with a temperature independent R_s or with a R_s having the temperature dependence given through the magnetic resistivity. Other parameters included in R_s, like the Fermi energy, carrier concentration and coupling constants like the normal exchange and spin-orbit coupling are considered temperature independent [38].

As shown in *iii*, it is not possible to describe the R_H of some Yb compounds with a temperature independent R_0 and R_s in (1). The required temperature dependence for R_s cannot be given through $\rho(T)$ either, because of the saturation (Fig. 1). By matching ΔR_H in a form of a constant times X or $(\rho-\rho_{ph})X$ at room temperature the values obtained are too low, i.e. to explain the experimental results above T_{ch}, $R_s(T)$ should increase with decreasing temperature.

4.2. *Comparison with the Theory*

A theory to explain the Hall anomalies of intermediate valent and Kondo compounds was recently proposed by Ramakrishnan et al. [3, 4]. As in the semiphenomenological theory of Newns and Hewson [36] the IV compounds are described as a collection of weakly interacting $4f$ resonances characterized by two parameters: The position of the resonance above the Fermi level ε_f and its width Δ. The Hall anomaly ΔR_H, according to the authors, is produced by a resonant skew scattering of the conduction electrons at the narrow $4f$ resonances splitted by the magnetic field, ΔR_H being proportional to a coupling constant $K(T)$ times X. For a Kondo regime at high temperatures ($T>\Delta^*$) $K(T)$ is proportional to $(1-\tilde{X}T)$, with $X=1/3(g\,\mu_B)^2\,J(J+1)\,\tilde{X}$. Although the assumption of such a $K(T)$ for most of the measured Yb compounds may lead to inconsistencies with other physical properties, the comparison of the experimental results reported here with the expression (9) in [4] for $T>T_{ch}$ renders some interesting features in agreement with the experiment.

We have:

$$R_H \cong R_0+\rho_b(g\,\mu_B)/N(1-\tilde{X}T)\,\tilde{X}\,(\text{sign Yb?}) \qquad (3)$$

For Yb^{3+} is $N=8$, $g=8/7$ and $J=7/2$.

ρ_b is the bulk resistivity of the concentrated impurity system. As there is no explicit calculation for Yb given in [4] the expected sign for the extraordinary contribution is not known.

To compare (3) with experimental results it is necessary to know R_0: as done in (2) we set $R_0=$constant. This assumption is justified if the proposed extraordinary contribution fits the temperature dependence of the measured R_H. To estimate R_0 we follow the usual procedure: by extrapolating the measured R_H vs $(1-\tilde{X}T)\,\tilde{X}$ for $\tilde{X}\to 0$. In the temperature region where (3) fits the experiment a straight line is obtained. The values of R_0 are shown in Table 1 (third column), being consistent with those obtained empirically and rendering also an estimate of R_0 for $YbAl_3$. Using these values for R_0 and the susceptibilities shown in Fig. 5 it is possible to calculate R_H with (3).

Curve a_1 (Fig. 2) shows the calculated R_H for YbCuAl setting $\rho_b=\rho(T)$. A fairly good qualitative and quantitative agreement with the experiment is obtained. By setting $\rho_b=\text{constant}=\rho(270\text{ K})=115\ \mu\Omega$ cm, the broken curve b_1 (Fig. 2) is obtained, which fits the experiment very well for $T>60$ K. The Hall coefficient calculated for $YbCu_2Si_2$ using $\rho_b=\rho(T)$ is given by the curve a_2 (Fig. 2). A qualitative and quantitative agreement with the measured R_H is achieved only above 160 K. Using as before a constant $\rho_b=\rho(270\text{ K})=115\ \mu\Omega$ cm in (3), we obtain the broken curve b_2, which gives a good fit to the experiment for $T>70$ K. For $YbInAu_2$ with $\rho_b=\rho(T)$ we obtain the experimental results only for $T>270$ K. If we use as before a constant $\rho_b=\rho(270\text{ K})$ it is only possible to match the experimental R_H for $T>200$ K, the calculated values below this temperature being significantly smaller than those measured. For $YbAl_3$ the qualitative and quantitative disagreement between the calculated and measured R_H is even larger, however if we use a constant $\rho_b=102\ \mu\Omega$ cm in (3) we obtain the curve b_3 (Fig. 7-I) which fits the experimental R_H for $T>90$ K. The ρ_b (T) required to fit exactly the experiment is shown in Fig. 7-II: it should be almost temperature independent for $T>50$ K and much bigger than the measured also shown in the same picture.

For YbPd formula (3) predicts a small anomaly for $T>150$ K in agreement with the experiment. As the Hall anomaly in this case is so small (Fig. 4), YbPd cannot give a reliable test for the validity of (3)

We see that the agreement between R_H calculated with (3) using $\rho_b = \rho(T)$ and the R_H measured deteriorates when going to compounds with increasing intermediate valence character, from Yb^{+3} to Yb^{+2}. It is nevertheless remarkable that the term $(1-\tilde{X}T)\,\tilde{X}$ with a constant ρ_b of about 110 μΩ cm fits the experimental R_H above a temperature consistent with the T_{ch} estimated from susceptibility measurements (see Table 1) and values of the parameter Δ given in [36], $YbInAu_2$ being the only exception. A plot of $((1-\tilde{X}T)\,\tilde{X})^{-1}$ vs T for YbCuAl and $YbCu_2Si_2$ renders straight lines with positive temperature extrapolations in agreement with Fig. 6. As mentioned in Sect. 4.1 this feature could not be explained with a temperature independent R_S.

The comparison of the measured R_H with the R_H values calculated with (3) shows that for YbCuAl and $YbCu_2Si_2$, two compounds with an almost integral valence, the temperature dependence of R_s for the high temperature regime may be given through the susceptibility in the form of a coupling constant $K(T)$ proportional to $(1-\tilde{X}T)$. It remains an open question whether other forms of the coupling constant are also suitable. In general, when the form (1) is assumed and when all parameters in R_H except $K(T)$ are considered as temperature independent, $K(T)$ above T_{ch} must increase with decreasing temperature to fit the experiment. The question arises whether such a temperature dependence of $K(T)$ would not produce a negative temperature coefficient in $\rho(T)$, a feature only shown by YbCuAl and YbPd when substracting the phonon resistivity of a reference.

There are some other aspects worth being considered.

The expression (3) does not take into account crystal field effects and it is not known if their inclusion would be essential to explain the Hall effect of Yb compounds.

It remains unknown why YbCuAl and $YbCu_2Si_2$ show anomalies with opposite signs. We may speculate that as the theory fits better YbCuAl, the predicted "ideal" sign for almost integral valent Yb ions should then be negative i.e. opposite to the sign predicted for Ce in [4].

With respect to the relationship between ρ_b and ΔR_H given by (3) we find a conflicting qualitative and quantitative mismatch between the measured resistivity ρ and ρ_b in (3), excepted for YbCuAl (curve a_1 in Fig. 2). As mentioned before the required ρ_b should be larger than ρ which is a contradiction. Curiously for $YbAl_3$ (see curve b_3 in Fig. 7-II) and $YbCu_2Si_2$ the $\rho_b(T)$ required to fit the experiment with (3) should be quantitatively and qualitatively similar to $\rho(T)$ for YbCuAl (see Fig. 1) i.e. temperature independent above some temperature with a steep decrease below it. For $YbCu_2Si_2$ and $YbAl_3$ this temperature corresponds to the inflexion point were the measured ρ settles into ρ_0 which may then be interpreted as a point where a drastic reduction of the asymmetric scattering occurs.

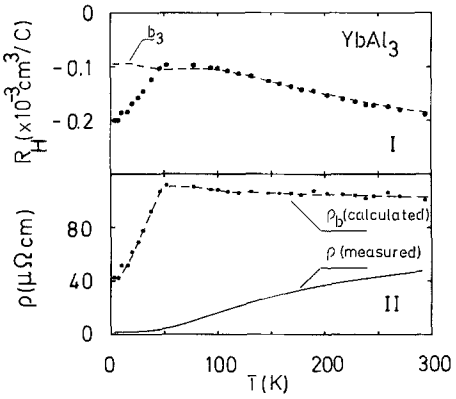

Fig. 7. I. Temperature dependence of the measured Hall coefficient R_H for $YbAl_3$. The curve b_3 shows the calculated R_H with $\rho_b =$ constant in the expression (3) of Sect. 4.2. II. Temperature dependence of the electrical resistivity (measured) and ρ_b (calculated with the expression (3) using the measured R_H)

These considerations give arise to the question of the applicability of (3) and of the basic assumptions of the theory to compounds with high valency and high characteristic temperatures like $YbAl_3$ or $YbInAu_2$. In a strict sense the only compound that follows (3) is YbCuAl. It is anyway a remarkable fact that a constant ρ_b of 110 μΩ cm in (3) can fit the experiment (curves b_1 and b_2 in Fig. 2 and b_3 in Fig. 7) above a certain temperature.

To explain the low temperature behavior ($T > \Delta$) in the frame of the theory an expression including X and the phase shifts δ_2 and δ_3 should be necessary. There are however some experimental features which indicate that this might not be sufficient. The theory assumes uncorrelated $4f$ resonances for the compounds. It is believed [36, 39] that the Yb compounds should suit this picture better than the Ce compounds because of their higher degeneracy ($N=8$). In fact the Yb compounds do not show the mysterious sign changes in R_H at low temperatures followed by strong temperature dependence of some prominent Ce compounds like $CePd_3$ [1, 2] $CeBe_{13}$ [2], or $CeCu_6$ [40] recently attributed to coherence effects [41]. The question is, whether some evidence exists for intercorrelations between the Yb ions in the present compounds.

The observed plateaus and maxima in R_H (Fig. 2) may be related to this, as suggested by certain similarities to those observed in spin-glass systems [42]

where they coincide to the on-set of a cooperative effect.

In the particular case of Yb IV compounds where the interaction of the $4f$ ions with the conduction electrons plays an important role for the stability of the magnetic moments, a transition from a regime characterized by an on-site hybridiazation ($T > \Delta$) to a regime dominated by coherent hibridization ($T < \Delta$), as suggested by many authors [9, 20, 36, 39], should become apparent in the Hall effect.

We believe that the steep decrease of R_H in the temperature interval where ρ settles into ρ_0 is relevant in this sense (see Sect. 1). If the elastic scattering producing ρ_0 does not contribute to ΔR_H (see Sect. 4.1) the main scattering source for skew-scattering should be some low energy excitations condensing at low temperatures. They would contribute low angle scattering causing the asymmetric part of the scattering probability to approach zero (Eq. (6) in [4]). The usual low energy excitations for low temperatures are long wavelength-longitudinal phonons which produce low angle scattering for $T < \theta_D/10$ (θ_D = Debye temperature). θ_D for YbCuAl, $YbCu_2Si_2$ and $YbInAu_2$ lies between 200 K and 300 K [18] coinciding with the temperature range where the steep decrease of R_H occurs. However for $YbAl_3$ the steep decrease at 50 K would predict a very high Debye temperature (no values of θ_D where found in the literature to check this). We believe that the influence of the phonons in the asymmetric scattering is only indirect.

It is also possible that the onset of a strong coherent hybridization at low temperatures produces simultaneously a more translationally invariant lattice thus reducing the scattering ($\rho \to \rho_0$) and the $4f$ character (quantum numbers J and L) of the ion. This reduces drastically the skew-scattering (we always assume that the asymmetric scattering is due to local moments).

The interpretation of the experimental results, so far, was given in terms of an extraordinary Hall effect, which we believe is the hypothesis which may be tested in a simpler way from the experimental point of view than any speculation about the normal Hall effect. It is shown by the experiment (see Sect. 3) and is consistent with the theory that Yb IV compounds with small susceptibilities ($< 4 \times 10^{-3}$ emu/mole) and high valencies like $YbAl_2$, Yb_3Bi_4 [43], Yb_3Sb_4 [43], Yb metal [44] do not show an extraordinary Hall effect i.e. in these cases $R_H(T) = R_0(T)$ holds.

In compounds showing an extraordinary contribution in R_H the extrapolated values of R_0 are smaller in absolute value than the R_H's of the corresponding reference compounds (see Table 1). In compounds with small extraordinary contribution like $YbAl_3$ and $YbInAu_2$ a temperature dependent R_0 could help to explain the low temperature behaviour. Some remaining question in this sense is if $YbInAu_2$ undergoes a new reversal of sign at lower temperatures or stays positive. In the later case a reversal of sign in $R_0(T)$ from negative to positive could be assumed. A similar situation is valid for YbCuAl.

4.3. Final Remarks

The analysis of the Hall effect measurements on Yb intermetallic compounds shows that the observed anomalies may be attributed to an extraordinary contribution to the Hall coefficient R_H. This skew scattering contribution dominates the temperature dependence of R_H in intermediate valent compounds having a valence state close to the Yb^{+3} configuration, and it decreases with increasing valence, i.e. for compounds with a larger Yb^{2+} mixing.

The fact that it is possible, above a characteristic temperature T_{ch}, to explain qualitatively the temperature dependence of the extraordinary contribution with the theory of Ramakrishnan et al. gives new support to the independent scatterer picture, providing the study of $R_H(T)$ an useful tool for testing different forms of the $c-f$ coupling mechanisms.

The low temperature behaviour of $R_H(T < T_{ch})$ is not so complex like the one observed for some Ce compounds [2, 37]. Its detailed interpretation requires, however, a more elaborate picture than the one used for $T > T_{ch}$ possibly including crystal fields effects and $f-f$ correlations.

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