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Skew scattering by rare-earth impurities in silver, gold, and aluminum*

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We report Hall-effect measurements on silver, gold, and aluminum containing rare-earth impurities, between 1.2 and 77 K and up to 40 kG. The Hall effect due to skew scattering by the magnetic rare-earth impurities is clearly observed; for 0.5 at.% of impurities and at 1.2 K, it can be twice as large as the ordinary Hall effect. We present a model which can explain the variation of the skew scattering throughout the rare-earth series and, from the analysis of the experimental results, we derive the coupling between the rare earth and the orbital angular momentum of a conduction electron.

I. INTRODUCTION

Skew scattering is known to be one of the mechanisms of the extraordinary Hall effect in ferromagnetic metals and was first discussed by Smit.[1] In recent years the problem of skew scattering by isolated magnetic atoms in nonmagnetic metals has also started to attract attention.[2-6] Clear experimental evidence of skew scattering by rare-earth (RE) impurities in silver has been provided by Hall-effect measurements that we have reported recently.[6] In order to understand more precisely the mechanism of skew scattering by RE impurities we have proceeded with a systematic study of the Hall effect at low temperature (1.2–77 K) of Ag:*R*, Au:*R*, and Al:*R* alloys. We present in this paper this experimental study (Secs. II–III) together with the theory of skew scattering that we propose (Sec. IV).

The scattering of conduction electrons by a magnetic impurity is called skew if the deflection is different to the right and to the left (with respect to the direction of the impurity magnetic moment). When the magnetic impurities are polarized by an applied field, they all deflect the electrical current in the same direction and so contribute to the Hall effect. One expects the Hall resistivity to be the sum of the following:

(a) A term ρ_{xy}^0 of ordinary Hall resistivity which is due to the Lorentz force. Under the condition that $\omega_c\tau \ll 1$ and the phonon resistivity is much smaller than the impurity's resistivity, this term is generally proportional to the magnetic field and independent of the temperature (and also independent of the impurity concentration in the dilute limit). This behavior of the ordinary Hall resistivity is observed, for instance, for silver with nonmagnetic impurities[7] when the conditions above are fulfilled. In dilute magnetic alloys the so called "spin effect"[8] may increase the ordinary Hall coefficient at high field but leaves the low-field Hall coefficient unchanged. Anyhow the spin effect is clearly related to the negative magnetoresistance and, if one considers the very weak negative magnetoresistance of Ag:*R* or Au:*R* alloys,[9,10] it should be quite ineffective in these systems. Actually we could not observe any spin effect in our Hall-effect measurements on Ag-, Au-, or Al-based alloys.

(b) A term ρ_{xy}^E (extraordinary Hall resistivity) due to the skew scattering; ρ_{xy}^E is proportional to the concentration of the magnetic impurities, proportional to their polarization in the low-field limit (and to H/T if the susceptibility follows a Curie law) and becomes saturated at high field (generally for $gJ\mu_B H \gg kT$). ρ_{xy}^E may be exactly proportional to the magnetization of the impurities at any field for some mechanisms of skew scattering but can depend on the field in a more complicated way.[4,5] In addition to the skew scattering, the mechanism of "side jump"[11,12] may also contribute to the extraordinary Hall resistivity but always for more concentrated alloys than those we have studied.

The scattering by a magnetic impurity can be skew if the matrix elements of the scattering potential between states of wave vector $\vec{k}$ and $\vec{k}'$ are changed by an interchange of $\vec{k}$ and $\vec{k}'$. Thus the exchange interaction between conduction and *f* electrons cannot make the scattering skew if it is simply written, as usual,

$$H = -\sum_{\vec{k}\vec{k}'} N^{-1} a_{\vec{k}'}^{*} a_{\vec{k}} (g-1)\,\Gamma(|\vec{k}-\vec{k}'|)\,\vec{s}\cdot\vec{J}, \qquad (1)$$

where $\vec{s}$ is the spin of the conduction electron, $\vec{J}$ the total angular momentum of the *f* electrons, and $(g-1)$ the de Gennes factor. However, when the localized electrons possess an orbital angular momentum, the exchange interaction cannot be reduced to a simple spin-spin interaction such as (1) but contains additional terms and, in particular, terms which couple the orbital angular momentum of the conduction electron $[\vec{l} = (1/\hbar)$

$\vec{r}\times\vec{p}]$ with $\vec{J}$.[13,14] We refer chiefly to the work of Kondo[14] who, limiting the plane waves of the conduction electrons to their s and p partial waves, derived a relatively simple form of the exchange interaction between conduction and f electrons [Eq. (2.45) in the Kondo paper[14]]. If one keeps only the terms relevant to the skew scattering problem, this interaction can be written

$$H_{cf} = -\sum_{\vec{k}\vec{k}'} N^{-1} a^{*}_{\vec{k}'} a_{\vec{k}} \{ (g-1)\,\Gamma(|\vec{k}-\vec{k}'|)\vec{J}\cdot\vec{s} + \tfrac{1}{2} i (2-g) \times F_2 \vec{J}\circ(\vec{\kappa}\times\vec{\kappa}') + 2id_2F_3[\vec{J}\cdot(\vec{\kappa}\times\vec{\kappa}')]\,(\vec{J}\circ\vec{s})\} \,, \tag{2}$$

where $\vec{\kappa}$ ($\vec{\kappa}'$) denotes the unit vector in the direction of $\vec{k}$ ($\vec{k}'$). The first term of (2) is the usual spin-spin exchange interaction, the second and third terms contain $(\vec{\kappa}\times\vec{\kappa}')$ in first order and so are antisymmetric terms which can give rise to skew scattering. One notes that, when the expansion of plane waves is limited to s and p partial waves, the matrix elements of the orbital angular momentum $\vec{l}$ between plane waves $\vec{k}$ and $\vec{k}'$ is $12\pi i(\vec{\kappa}\times\vec{\kappa}')$, so that the second and third terms correspond to interactions of the type $\vec{J}\cdot\vec{l}$ and $(\vec{J}\cdot\vec{l})(\vec{J}\cdot\vec{s})$ but restricted to the p partial waves. Only the second term will be considered in the calculation of Sec. IV. Appendix B shows that the third term induces a much smaller contribution to the Hall effect.

The Hall resistivity arising from the term of the type $\vec{J}\circ\vec{l}$ in H_{cf} has been already calculated by Giovannini[4] and will be again calculated in Sec. IV in a somewhat different way. This contribution to the Hall resistivity cancels out for Gd (S-state ion, $g=2$) as the coefficient of $\vec{J}\cdot(\vec{k}\times\vec{k}')$ in (2) cancels for $g=2$. However the experimental results show that the skew scattering is maximum for Gd in Ag or Au. It turns out that there should be in these alloys a second mechanism of skew scattering which does not arise from the asymmetry of H_{cf}. We show in Sec. IV that the admixture of the conduction electrons with the nonmagnetic $5d$ electrons of the RE can also give rise to skew scattering if the spin-orbit coupling of the $5d$ electrons is taken into account. A combination of this mechanism and of the preceding mechanism based on the $(\vec{l}\cdot\vec{J})$ term of H_{cf} can explain the experimental results and, in particular, the variation of the skew scattering throughout the RE series. This is discussed in Sec. V. The fit with the experimental data allows us to obtain interesting information on the magnitude of the term $\vec{l}\cdot\vec{J}$ in the interaction between the conduction and f electrons.

II. EXPERIMENTAL PROCEDURE

The silver- and gold-based alloys have been mostly prepared at the University of Geneva by arc melting with water-cooled copper hearth and tungsten electrode. The gold and silver metals were obtained from Engelhard Industries and were nominally 99.999% pure. The rare-earth metals were obtained from Nuclear Corporation of America and were 99.9% pure.

The aluminum-based alloys and some silver and gold alloys with light RE were prepared at the CEN Saclay by melting in a levitation furnace and then quenched by splat cooling.

The arc-melted gold and silver alloys were cleaned in aqua regia for gold alloys or nitric acid for silver alloys, rolled into foils of thickness 0.1 mm, and cleaned again to obtain a thickness of 0.07 mm. The foils were accurately cut by spark machining to the shape for Hall-effect measurements, then annealed 5 h at 800 °C in sealed quartz tubes containing argon and quenched in water.

This thermal treatment appeared to be the best to obtain the maximum solubility, as it has already been found by Bijvoet *et al*.[15] for the Ag:*R* alloys. These authors have performed a systematic study of the solubility of the RE in silver. They have measured the residual resistivity per at.% in the very dilute limit ($c\sim 500$ ppm), observed that, when the concentration is raised, the beginning of the RE clustering results in a decrease of the resistivity per at.% and determined the limit of solid solubility in this way. Above the limit of solubility the resistivity is almost independent of the concentration and this shows that the resistivity per atom is much smaller for clustered than for isolated impurities. We also observed for the Ag and Au based alloys a proportionality of the resistivity to the concentration at low concentration, followed by a saturation of the resistivity above the limit of solubility. Moreover, we purposely clustered the RE of some alloys by an annealing at 400 °C without quenching and observed then an almost complete cancellation of the resistivity and, at the same time, of the skew-scattering term. This confirms that the clustered atoms contribute weakly to the normal and also to the skew scattering. The resistivity defines nearly the effective concentration of isolated impurities.

Table I shows the nominal concentrations, the residual resistivities and the effective concentrations of the Ag- and Au-based alloys studied in

TABLE I. Nominal concentrations, residual resistivities, and effective concentration (see text) of the Ag- and Au-based alloys studied in the paper.

	Ag-based alloys			Au-based alloys		
Rare-earth element	Nominal concentration (at.%)	Residual resistivity ($\mu\Omega$ cm)	Effective concentration (at.%)	Nominal concentration (at.%)	Residual resistivity ($\mu\Omega$ cm)	Effective concentration (at.%)
Pr	0.5	0.36	0.13	0.5	0.96	Not determined
Nd	0.5	0.5	0.14	0.5	0.46	...
Sm	0.5	1.1	0.29	1	1.2	...
Gd	1	3.76	0.71	0.5	3.4	0.5
Tb	0.5	3.54	0.56	1	6.84	0.98
Dy	0.3	1.8	0.27	1.5	9.81	1.40
Dy	0.4	2.2	0.33	0.5	3.52	0.5
Dy	0.8	5.5	0.82	1	7.21	1.03
Dy	1	6.67	1			
Dy	1	7.07	1.06			
Ho	1	5.89	0.96	1	6.9	0.99
Er	2	11.8	1.95	1	6.40	0.91
Tm	2	11.8	2.0	1	6.04	0.86
Yb				2	14.3	2.04
Lu	1	6.1		1	6.21	0.89

this paper. For the Ag-based alloys, the effective concentrations are obtained by dividing the residual resistivity by the resistivity per at.% found by Bijvoet *et al.*[15] in the dilute limit. For the gold-based alloys measurements by Edwards and Legvold[16] or Murani[17] indicates that the resistivity per at.% is about 7$\mu\Omega$cm for most of the heavy RE and we adopted this value throughout the series to calculate the effective concentration. It can be seen in Table I that, for heavy RE, 0.5 at.% and sometimes 1 or 2 at.% could be dissolved in Ag or Au (one obtains a good agreement between the nominal and effective concentration). On the contrary it has been impossible to dissolve completely 0.5 at.% of light RE and the residual resistivities obtained correspond to smaller effective concentrations. A part of the RE is then certainly clustered but, as we have seen above, it should contribute very weakly to the electron scattering.

The aluminum based alloys have been obtained by splat cooling. Only Tb, Dy, Ho, Er could be dissolved at sufficient concentration for the observation of skew scattering ($c \sim 1000$ ppm). We have some reasons to believe that clusters (RE or Al:R compounds) are present in the alloys. A very approximate concentration was determined by comparing the resistivity with that of very dilute alloys.[18] The splat-cooling technique was also used for some gold-based alloys. We have checked that the splat cooling introduces only a negligible defect scattering.

The Hall-effect measurements were performed in a superconductive coil up to 40 kG and from 1.2 to 77 °K. The current in the sample was modulated at 22 Hz and the Hall voltage was detected by a lock-in PAR amplifier (sensitivity of the measurement: $\pm 10^{-9}$ V). Details on the apparatus have been already reported.[19]

III. EXPERIMENTAL RESULTS

The main features of the Hall resistivity induced by skew scattering have been described in Sec. I and it appears that the skew scattering shows up first in the temperature dependence of the Hall effect. On Figs. 1–5 we have plotted the low-field Hall coefficient R (R is the limit of ρ_{xy}/H when $H \to 0$) versus T^{-1} for series of Ag:R and Au:R alloys.

We consider first the plots for *Ag*Gd on Fig. 1 and *Au*Gd on Fig. 3: it turns out that R can be decomposed in the sum of a constant term R_0 (the ordinary Hall coefficient of the *Ag*Gd or *Au*Gd alloys) and of a term AT^{-1} that we ascribe to skew scattering by the Gd impurities

$$R = R_0 + AT^{-1}. \tag{3}$$

A skew-scattering term varying in T^{-1} is just what is expected for Gd impurities whose initial susceptibility obeys a Curie law. A constant value of the ordinary Hall coefficient is quite normal as the impurity scattering remains much stronger than the phonon scattering throughout the temperature range.[7]

For the other magnetic alloys (e.g., *Ag*Tb on Fig. 1), a variation $R = R_0 + AT^{-1}$ is clearly observed at high enough temperature. At lower

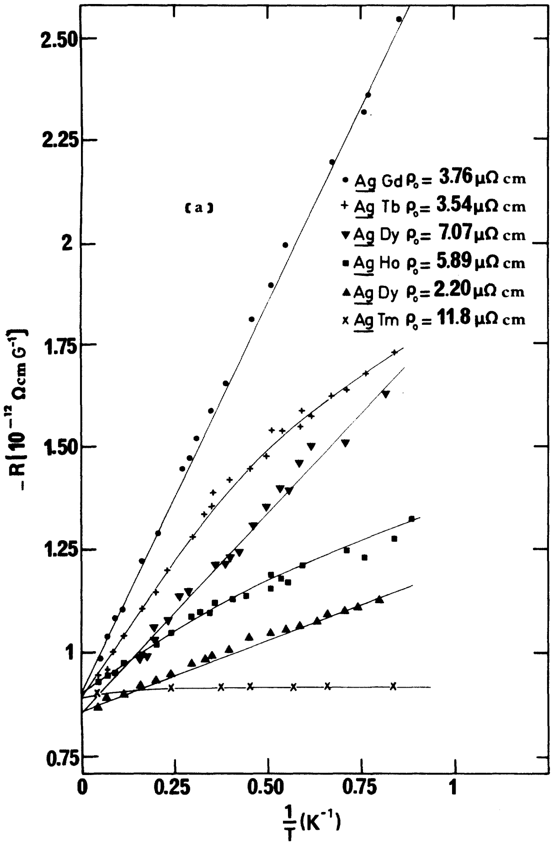

FIG. 1. Initial Hall coefficient (R is the limit of ρ_{xy}/H for $H\rightarrow 0$) vs T^{-1} for *Ag*Gd, *Ag*Tb, *Ag*Dy, *Ag*Ho, *Ag*Tm alloys.

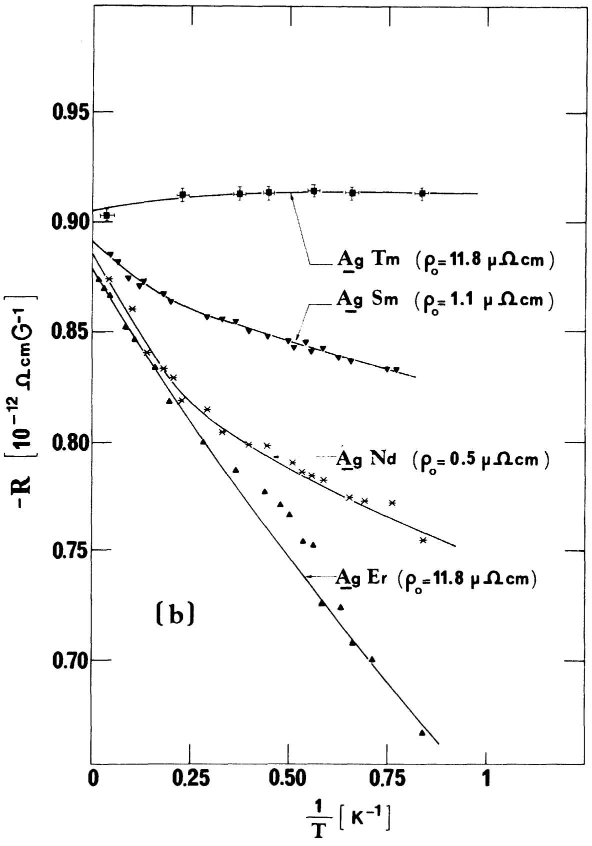

FIG. 2. Initial Hall coefficient R vs T^{-1} for *Ag*Tm *Ag*Sm, *Ag*Er, *Ag*Nd alloys.

temperature one observes deviations from T^{-1} which can be explained by crystal-electric-field (CEF) effects. Actually the initial magnetic susceptibility[17,20] of most of the alloys also shows at low-temperature deviations from the Curie law observed at high temperature, and the deviations are explained by the splitting of the f level by the CEF. On Fig. 6 we have plotted at the same time the inverse of the magnetic susceptibility (from Murani[17]) and $(R-R_0)^{-1}$ as a function of the temperature for a *Au*Dy alloy. It appears that one can make the curves coincide, and this means that the skew-scattering term has the temperature dependence of the susceptibility.

In some gold alloys with a very small skew-scattering effect, the analysis of the temperature dependence of the Hall coefficient is not so simple as other contributions cannot be neglected. Considering the plot of R vs T^{-1} for the nonmagnetic alloy *Au*Lu on Fig. 4, one observes that R is not rigourously independent of the temperature (on the contrary R is quite flat for *Ag*Lu or *Ag*Tm on Fig. 1). We believe that the change of R in *Au*Lu may be due to 10 ppm of residual iron impurities (which are known to contribute significantly to the Hall effect of gold above[19] 5 °K). The effect of such residual iron impurities may be non-negligible for alloys which exhibit a very small skew-scattering effect, for instance *Au*Er, *Au*Tm, and *Au*Yb. It turns out that, for *Au*Er, *Au*Tm, and *Au*Yb, the skew-scattering term can be determined accurately only below 5 °K. For the *Au*Pr and *Au*Sm alloys, the dip of R above 15 °K on Fig. 5 is obviously not due to skew scattering, but may be due either to the presence of iron impurities or to a change of the ordinary Hall effect as, in these low-concentrated alloys, the phonon scattering is not quite negligible above 15 °K. These several alloys (*Au*Er, *Au*Tm, *Au*Yb, *Au*Sm, *Au*Nd) are those for which the analysis of the temperature dependence of the Hall coefficient is somewhat questionable. For

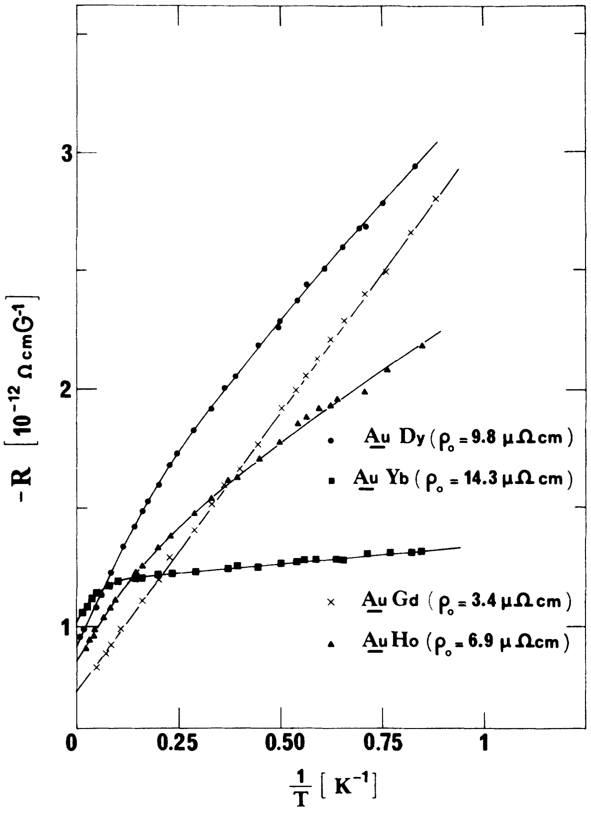

FIG. 3. Initial Hall coefficient R vs T^{-1} for *Au*Gd, *Au*Dy, *Au* Ho, *Au*Yb.

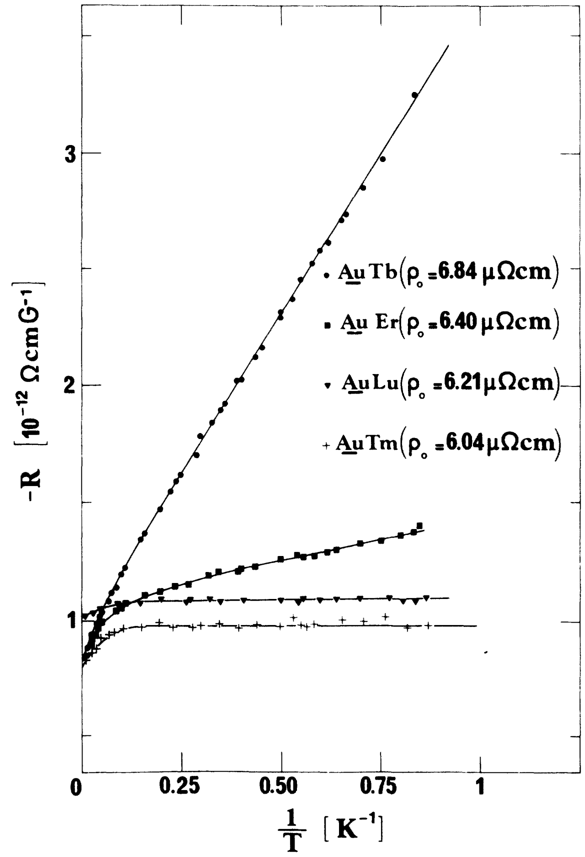

FIG. 4. Initial Hall coefficient R vs T^{-1} for *Au* Tb, *Au* Er, *Au* Tm, *Au* Lu.

the other alloys, the skew-scattering effect is much larger and can be determined much more accurately.

The Hall effect induced by skew scattering is also expected to be proportional to the impurity concentration. We have determined the coefficient A of the term in T^{-1} in the Hall coefficient of several *Ag*Dy and *Au*Dy alloys at high temperature and plotted A as a function of the concentration C and of the residual resistivity ρ_0 on Figs. 7 and 8. It can be seen that A is proportional to C or ρ_0. So, at least at high temperature, the experimental Hall coefficient can be written

$$R = R_0 + a\rho_0 T^{-1} . \qquad (4)$$

The magnitude of the skew scattering by a given rare-earth impurity in the high-temperature limit can be characterized by the coefficient a:

$$a = \left(\frac{R - R_0}{\rho_0} T\right)_{T^{-1}\to 0} = \left(\phi \frac{T}{H}\right)_{T^{-1}\to 0, H\to 0}$$

(ϕ means the Hall angle induced by the magnetic impurities).

On Figs. 9–11, the Hall resistivity ρ^E_{xy} induced by skew scattering in *Ag*Gd, *Au*Gd, and *Au*Tb alloys is plotted as a function of the magnetic field at several temperatures. To obtain ρ^E_{xy}, we determine the ordinary Hall coefficient R_0 of the alloy on the Figs. 1–5 and we subtract R_0H from the experimental Hall resistivity ρ_{xy}. We emphasize that the high resistivity of the alloys justifies us in taking the ordinary Hall resistivity linear in H ($\omega_c\tau \ll 1$) and independent of the temperature ($\rho_{\text{phonons}} \ll \rho_{\text{imp}}$ in the experimental temperature range, except for some low-concentrated alloys); this behavior is actually observed when the skew-scattering effect cancels out (*Ag*Tm, *Ag*Lu, *Au*Lu).

We consider first the variation of the Hall resistivity ρ_{xy} as a function of the field for the alloys with gadolinium impurities, for instance, *Au*Gd. On Fig. 9 we have also plotted Brillouin functions $B_{7/2}$ for T = 6.2 and 2 °K. One observes that, at

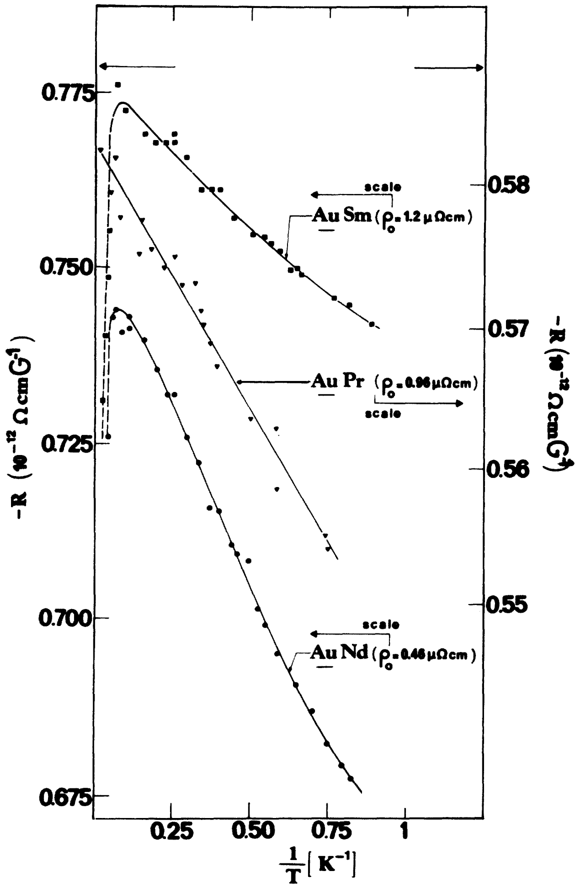

FIG. 5. Initial Hall coefficient R vs T^{-1} for *Au* Pr, *Au* Nd, *Au* Sm alloys.

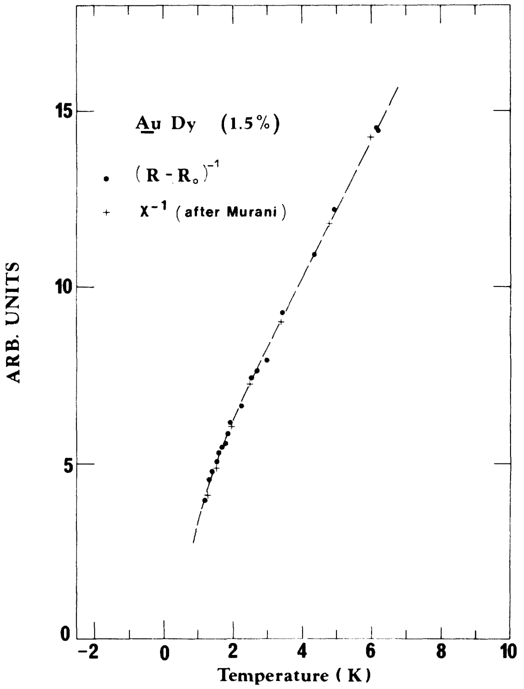

FIG. 6. Inverse of the magnetic susceptibility [from Murani (Ref. 17)] and $(R - R_0)^{-1}$ (R is the initial Hall coefficient, R_0 the ordinary Hall coefficient) vs T for Au + 1.5-at.% Dy.

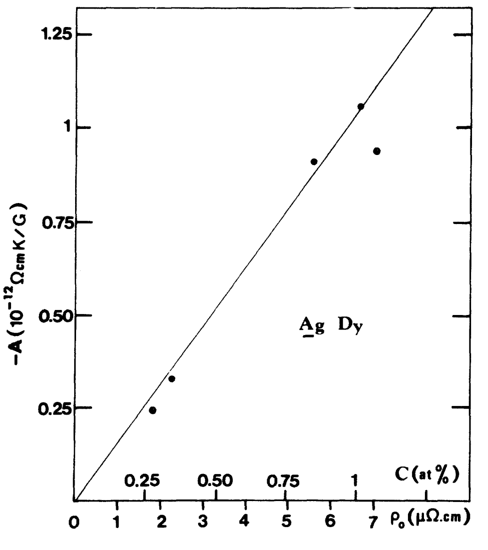

FIG. 7. A vs the residual resistivity and the concentration [A is defined by expression (3)] for *Ag* Dy alloys.

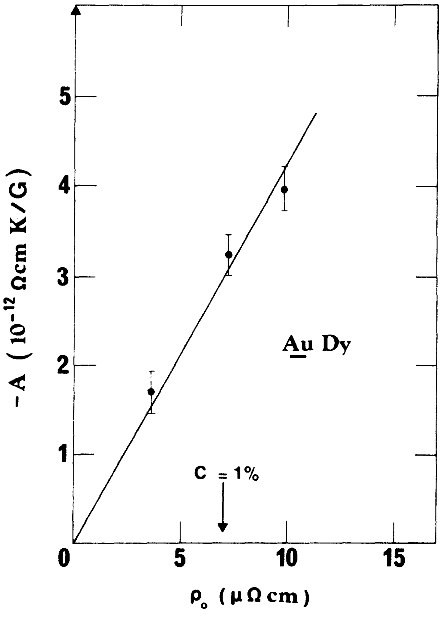

FIG. 8. A vs the residual resistivity for *Au* Dy alloys [A is defined by expression (3)].

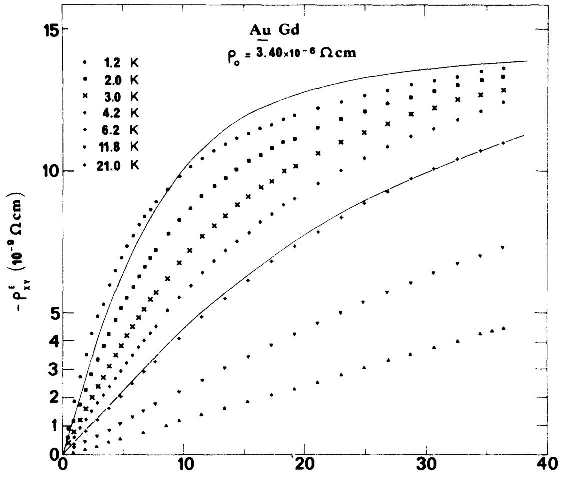

FIG. 9. Extraordinary Hall resistivity ρ^E_{xy} of a *Au*Gd alloy vs the magnetic field at several temperatures. The solid lines represent Brillouin functions for $J = \frac{7}{2}$, $g = 2$, $T = 6.2$ and 2 °K.

6.2 °K, the Hall resistivity ρ^E_{xy} induced by skew scattering varies very nearly like the corresponding Brillouin function. At 2 °K, ρ^E_{xy} departs somewhat from the corresponding Brillouin function. For *Ag*Gd also (Fig. 10) ρ^E_{xy} varies approximately like a Brillouin function. The theory described in Sec. IV predicts that the main skew-scattering contribution to the Hall resistivity is proportional to $\langle J_z \rangle$. This is in agreement with the observed fit with Brillouin functions. The slight deviations could be due to interaction effects at low temperature.

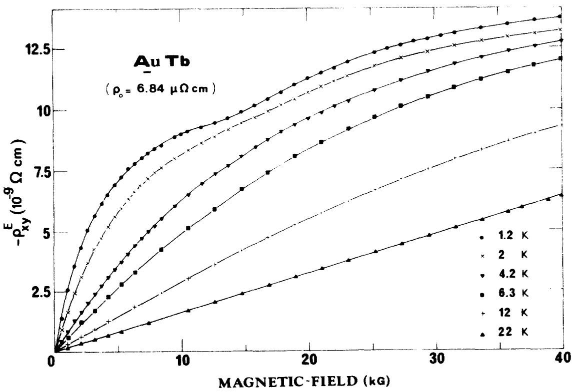

FIG. 11. Extraordinary Hall resistivity ρ^E_{xy} of a *Au* Tb alloy vs magnetic field at several temperatures.

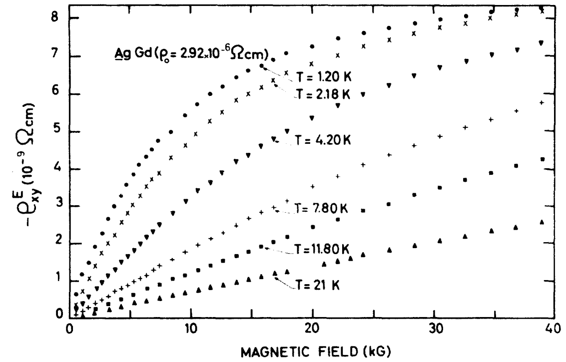

FIG. 10. Extraordinary Hall resistivity ρ^E_{xy} of a *Ag* Gd alloy vs magnetic field at several temperatures.

For impurities with CEF splitting, the field dependence of ρ^E_{xy} may be very different from that of a Brillouin function. The example of the *Au*Tb alloy is given in Fig. 11. If we consider the curves at low temperature, we observe a first beginning of saturation above 10 kG, followed by a rise at about 17 kG and again a saturation. Such complicated variations are observed for several non-*S* ions and are linked with the complex level structure in presence of crystal field and magnetic field of the same order of magnitude.

Although the analysis of the CEF effects on the field and temperature dependence of the skew scattering is interesting, it is not in our present scope and will be developed elsewhere. In this paper we want chiefly to clear up the origin and the mechanism of the skew scattering by RE impurities and, in order to avoid a special analysis of the CEF effects for each alloy, we shall only discuss in Sec. V the experimental data in the temperature range ($kT \gtrsim$ crystal field) where the magnetic susceptibility nearly reaches its free-ion value. In this range the experimental Hall coefficient (Figs. 1–5) can be fitted with expression (4), and the coefficient a characterizes the skew scattering for a given impurity. The experimental values of a are shown on Fig. 12 for the silver-based alloys and Fig. 13 for the gold-based alloys. For mostly of the alloys we have determined the coefficient a from experimental in the range 20–40 K. It can be checked from the magnetic measurements of Williams and Hirst[20] or Murani[17] that, down to 20 °K, the susceptibility deviates little[21] from the Curie law observed at higher temperature. In the case of *Au*Er, *Au*Tm, and *Au*Yb alloys, we could measure accurately the variation of the Hall coefficient only at low temperature (see above), and we determined the coefficient a after correction of the crystal-field effects.

We report finally the few results obtained for

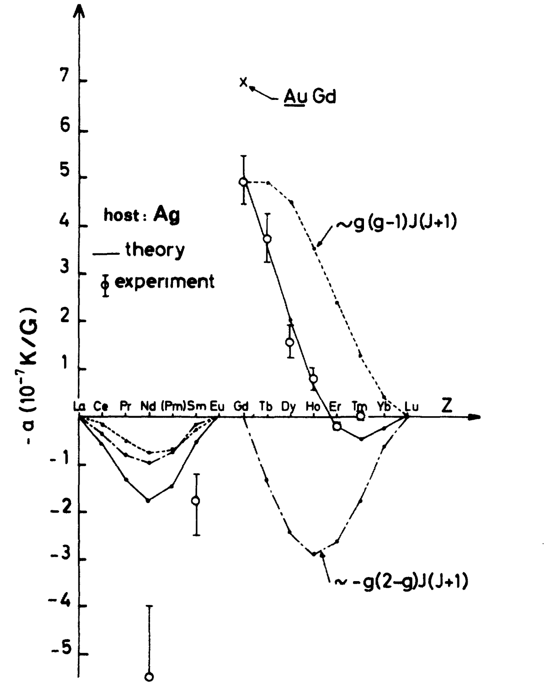

FIG. 12. Coefficient a which characterizes the skew scattering by each RE impurity in Ag is plotted throughout the RE series [a is defined by (4)]. The solid, dotted dashed, and dashed lines, represent, respectively, the values of a, a_1, a_2 calculated from (33) ($a=a_1+a_2$).

the Al:R alloys. On Fig. 14 the Hall constant of the Al:R alloys is plotted as a function of T^{-1}. The coefficients a derived from the experimental results are plotted on Fig. 15. The accuracy on these values of a is rather poor. We retain chiefly the sign of a and its rough variation in the RE series.

IV. THEORY

In order to derive the Hall resistivity induced by skew scattering, we have first to calculate the antisymmetric part of the scattering probability [antisymmetric in the sense $w^a(\vec{k}\to\vec{k}')=-w^a(\vec{k}'\to\vec{k})$]. It can be shown (see Appendix A) that antisymmetric terms in the scattering probability cannot arise in first Born approximation, so that the scattering probability must be calculated to second Born approximation. Secondly, these antisymmetric terms can only occur if two partial waves (with $l+l'=\text{odd}$) are scattered.[5] This requires a scattering potential acting on several partial waves and also complicates the calculation. We shall calculate the scattering probability of plane waves to second Born approximation with the following scattering potential of

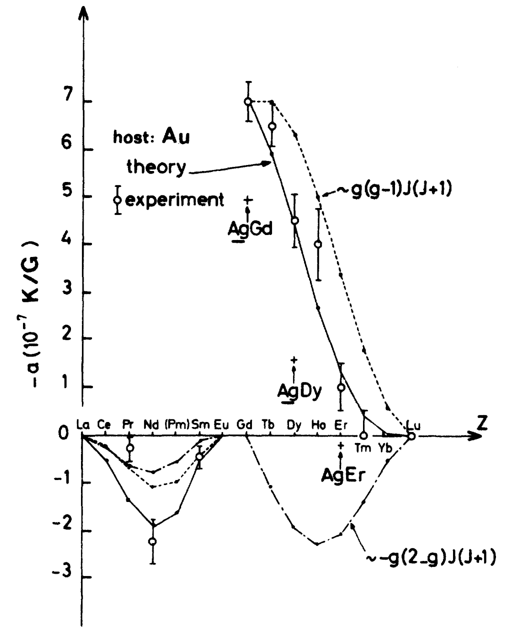

FIG. 13. Coefficient a which characterizes the skew scattering by each RE impurity in Au is plotted throughout the RE series [a is defined by (4)]. The solid, dotted-dashed, and dashed lines, respectively, represent the values of a, a_1, and a_2 calculated from (34) ($a=a_1+a_2$).

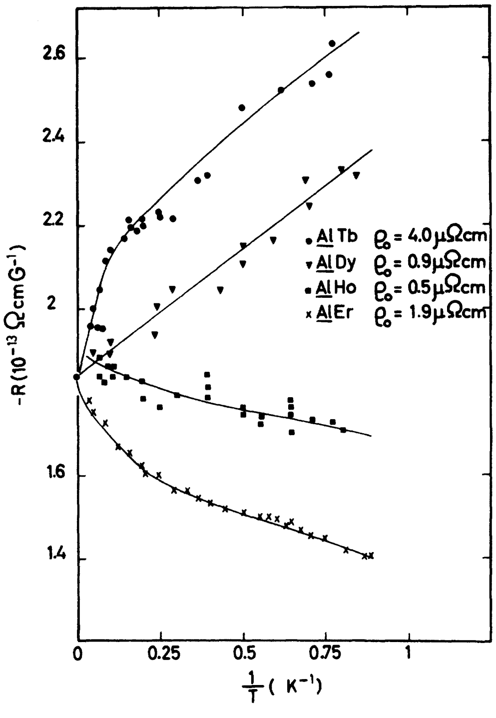

FIG. 14. Initial Hall coefficient R vs T^{-1} for AlTb, AlDy, AlHo, AlEr alloys.

the RE impurity:

$$H_S = H^{(1)} + H^{(2)} + H^{(3)} + H^{(4)},$$

where $H^{(1)}$ and $H^{(2)}$ are the exchange terms which couple the spin and the orbital angular momentum with the $4f$ moment, whereas the dominant terms $H^{(3)}$ and $H^{(4)}$ account for the attractive potential of a trivalent RE ion for d and s partial waves, respectively. We shall now specify the several terms of H_S and determine the corresponding scattering amplitude:

(i) $H^{(1)}$ is the usual spin-spin exchange interaction [first term of the interaction (2) derived by Kondo[14]]

$$H^{(1)} = -\sum_{\vec{k}\vec{k}'} N^{-1}(g-1)\Gamma(|\vec{k}-\vec{k}'|) \times \tfrac{1}{2}[(\overset{*}{a}_{\vec{k}'+}a_{\vec{k}+} - \overset{*}{a}_{\vec{k}'-}a_{\vec{k}-})J_z + \overset{*}{a}_{\vec{k}'+}a_{\vec{k}-}J^- + \overset{*}{a}_{\vec{k}'-}a_{\vec{k}+}J^+]. \tag{5}$$

Following Watson, Freeman, and Koide,[22] we write

$$\Gamma(|\vec{k}-\vec{k}'|) = 4\pi \sum_{l\leq 3,m} Y_{lm}^*(\Omega_{\vec{k}})Y_{lm}(\Omega_{\vec{k}'})\Gamma^{(l)}(|\vec{k}|,|\vec{k}'|), \tag{6}$$

$$\Gamma^{(l)}(k_F,k_F) = \Gamma^{(l)} . \tag{7}$$

The definition of the spherical harmonics Y_{lm} is that of Edmonds.[23] Throughout the paper their axis is that of the field. We limit the expansion to $l \leqslant 3$.

We show in Appendix B that it is sufficient to consider scattering without spin flip and to keep only the scattering amplitude terms of first order in $H^{(1)}$, that is,

$$f^{(1)}_{\vec{k}\pm}(\Omega_{\vec{k}'}, \pm) = \pm \frac{2\pi^2 n(E_F)(g-1)}{k_F} J_z \sum_{l,m} e^{2i\eta_l} \times \Gamma^{(l)} Y_{lm}^*(\Omega_{\vec{k}})Y_{lm}(\Omega_{\vec{k}'}), \tag{8}$$

where $n(E_F)$ is the one-spin density of states at the Fermi level in the conduction band [the factor $\exp(2i\eta_l)$ accounts for the phase shift of the partial waves $l=0$ and $l=2$ by the dominant attractive potential to which the small exchange is added].

(ii) $H^{(2)}$ is the second term of the general exchange interaction derived by Kondo.[14] We recall that this term correspond to a coupling of the type $\vec{l}\cdot\vec{J}$ (but limited to the partial waves $l=1$) between the orbital angular momentum $\vec{l}$ of the conduction electrons and the total angular momentum $\vec{J}$ of the f electrons. Only the part of $H^{(2)}$ which does not flip $\vec{J}$ is useful in our calculation (Appendix B). We write it in a form similar to (5):

$$H^{(2)} = -\sum_{\vec{k}\vec{k}'} N^{-1}(2-g)\tfrac{2}{3}\pi F_2 J_z \sum_m m \times Y_{1m}^*(\Omega_{\vec{k}})Y_{1m}(\Omega_{\vec{k}'})\overset{*}{a}_{\vec{k}'}a_{\vec{k}}. \tag{9}$$

It is sufficient to calculate the scattering amplitude arising from $H^{(2)}$ in first Born approximation (see Appendix B), and one obtains

$$f^{(2)}_{\vec{k}\pm}(\Omega_{\vec{k}'}, \pm) = \frac{2\pi^2 n(E_F)(2-g)F_2}{3k_F} J_z \sum_m m \times Y_{1m}^*(\Omega_{\vec{k}})Y_{1m}(\Omega_{\vec{k}'}) . \tag{10}$$

This term of the scattering amplitude is antisymmetric and can give rise to skew scattering. But it vanishes for Gd ($g=2$) and so we need another antisymmetric term to explain the experimental results for the alloys containing gadolimium impurities.

(iii) $H^{(3)}$ is the part of the attractive potential acting on the d partial waves. It is indeed well known that the substitution of a trivalent RE ion in a monovalent metal (Ag, Au) results in a potential capable of attracting about two conduction electrons, mostly in $5d$ states (formation of a $5d$ virtual bound state) and in $6s$ states. This is consistent with the values of the RE residual resistivities[15,16,26] which are much too high to be due only to the exchange scattering and which are roughly independent of the RE spin. And the crystal field seen by the $4f$ electrons can also be explained[20,24] by the formation of a $5d$ virtual bound state (vbs). For Al-based alloys

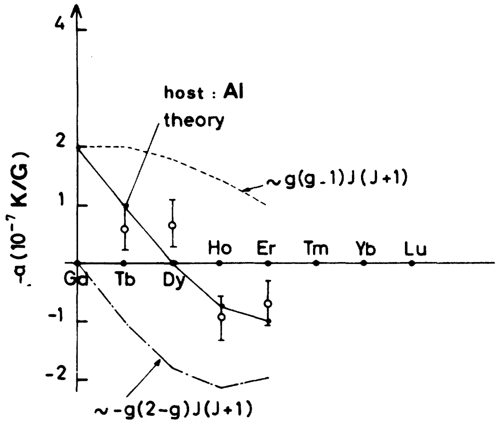

FIG. 15. Coefficient a which characterizes the skew scattering by each RE impurity in Al is plotted throughout the RE series [a is defined by (4)]. The solid, dotted-dashed, and dashed lines represent the values of a, a_1, and a_2 calculated from (35) ($a = a_1 + a_2$).

it has also been proposed[25] that the formation of a $5d$ vbs (by attraction of $5d$ electrons and repulsion of $3p$ electrons) could explain the residual g shift in the bottle-neck regime of EPR. The $5d$ vbs is generally supposed nonmagnetic as the experimental magnetic moments are nearly those of the trivalent ions (the small excess moment is explained by the polarization of the $5d$ vbs by the exchange coupling with the f electrons[26]).

The formation of a $5d$ vbs results in a strong scattering of the partial waves $l=2$. If the spin-orbit coupling of the d states is ignored, the phase shift of the partial waves $l=2$ is given by the classical expression[27]

$$\cot\eta_2 = (E_d - E)/\Delta, \tag{11}$$

where E_d is the energy of the center of the vbs and Δ its half-width. If the spin-orbit coupling of the d states is now taken into account,[28] the degeneracy of the states $j=\frac{5}{2}$ and $j=\frac{3}{2}(\vec{j}=\vec{l}+\vec{s})$ is raised and the vbs splits into two vbs centered at $E_{5/2}$ and $E_{3/2}$ with

$$E_{5/2} - E_{3/2} = \tfrac{5}{2}\lambda, \tag{12}$$

where λ is the spin-orbit constant of the $5d$ states $(E_{\text{SO}} = \lambda\vec{l}\cdot\vec{s})$. For simplicity we ignore the additional crystal-field splitting.

After Yafet[28] λ may be somewhat larger than the atomic spin-orbit constant as the orbit splitting lowers the correlation energy:

$$\lambda = \lambda_{5d}[1-(U-J)\rho_d]^{-1}. \tag{13}$$

However, for $5d$ electrons which are generally admitted to be far from the condition for magnetism, this enhancement should be small.

The splitting of the vbs results in phase shifts different for the partial waves of total angular momentum $\frac{5}{2}$ and $\frac{3}{2}$. As $\lambda\,(\lambda_{5d}\sim 10^{-1}\text{ eV})$ is smaller than $\Delta\,(\Delta\sim 0.2-0.6\text{ eV})$, the phase shifts $\eta_{5/2}$ and $\eta_{3/2}$ are obtained by differentiating (11). One obtains

$$\Delta\eta = \eta_{3/2} - \eta_{5/2} = \tfrac{5}{2}(\lambda/\Delta)\sin^2\eta_2, \tag{14}$$

where η_2 is the mean phase shift. We expand now the plane waves in partial waves of definite total angular momentum:

$$\begin{aligned}|\vec{k}_\pm\rangle = -4\pi j_2(kr)\sum_m Y^*_{2m}(\Omega_{\vec{k}})\Bigg[\left(\frac{3\pm m}{5}\right)^{1/2}|\,l=2, s=\tfrac{1}{2}, j=\tfrac{5}{2}, j_z=m\pm\tfrac{1}{2}\rangle\\ \mp\left(\frac{2\mp m}{5}\right)^{1/2}|\,l=2, s=\tfrac{1}{2}, j=\tfrac{3}{2}, j_z=m\pm\tfrac{1}{2}\rangle\Bigg]\\ +\text{terms with } l\neq 2.\end{aligned} \tag{15}$$

The asymptotic form $(r\to\infty)$ of the scattered wave function is obtained by introducing the phase shift $e^{i\eta_{5/2}}$ and $e^{i\eta_{3/2}}$ in the asymptotic form of the above expression and one derives straightforwardly the scattering amplitude. We need only the non-spin-flip scattering amplitude (see Appendix B) and we obtain

$$\begin{aligned}f^{(3)}_{\vec{k}\pm}(\Omega_{\vec{k}'},\pm) = \frac{2\pi}{ik_F}\sum_m\bigg((e^{2i\eta_{5/2}}-1)\frac{3\pm m}{5}Y^*_{2m}(\Omega_{\vec{k}})\times Y_{2m}(\Omega_{\vec{k}'})\\ +(e^{2i\eta_{3/2}}-1)\frac{2\mp m}{5}Y^*_{2m}(\Omega_{\vec{k}})\,Y_{2m}(\Omega_{\vec{k}'})\bigg).\end{aligned} \tag{16}$$

When $\eta_{5/2}$ and $\eta_{3/2}$ are different, the scattering amplitude $f^{(3)}$ is not symmetric with respect to $\vec{k}$ and $\vec{k}'$. An expansion to the first order in $\Delta\eta$ allows to separate the symmetric part $f^{(3s)}$ and the antisymmetric part $f^{(3a)}$:

$$\begin{aligned}f^{(3s)}_{\vec{k}\pm}(\Omega_{\vec{k}'},\pm) = \frac{2\pi}{ik}\sum_m(e^{2i\eta_2}-1)Y^*_{2m}(\Omega_{\vec{k}})\\ \times Y_{2m}(\Omega_{\vec{k}'}),\end{aligned} \tag{17}$$

where η_2 is the mean phase shift. This is the usual expression of the scattering amplitude when the spin-orbit coupling is ignored. The antisymmetric part is

$$\begin{aligned}f^{(3a)}_{\vec{k}\pm}(\Omega_{\vec{k}'},\pm) = \mp\frac{4\pi}{5k_F}\Delta\eta e^{2i\eta_2}\sum_m mY^*_{2m}(\Omega_{\vec{k}})\\ \times Y_{2m}(\Omega_{\vec{k}'}).\end{aligned} \tag{18}$$

Appendix B shows that the main contribution to the Hall effect is derived from the real part of $f^{(3a)}$. From (18) we write

$$\begin{aligned}f^{(3a)}_{\vec{k}\pm}(\Omega_{\vec{k}'},\pm) = \pm\frac{2\pi}{ik_F}\frac{\lambda}{\Delta}\sin^2\eta_2\sin 2\eta_2\sum_m mY^*_{2m}(\Omega_{\vec{k}})\\ \times Y_{2m}(\Omega_{\vec{k}'}) + \text{imaginary}.\end{aligned} \tag{19}$$

Thus the splitting of the nonmagnetic vbs by the spin-orbit coupling introduces in the scattering amplitude an antisymmetric term which has the opposite sign for the spin-up and spin-down electrons. We emphasize that this term should be nearly independent of the RE element as the $5d$ screening and the width of the vbs should be nearly equal for all the RE impurities in a given host.

The scattering amplitude $f^{(3)}$ correspond to a scattering potential which includes, in addition to a spin-independent potential V_2 acting only on the d partial waves, a term $\lambda' \vec{1} \cdot \vec{s}$ acting also on the d partial waves with λ' given by

$$\lambda' = \lambda \frac{\sin^2\eta_2}{2\pi\Delta\; n(E_F)}. \tag{20}$$

This term may be significant if $\sin^2\eta_2$ is of the order of the unity and if the vbs is narrow.

(iv) $H^{(4)}$ is the spin-independent potential which attracts the part s of the screening charge s. The corresponding scattering amplitude can be written

$$f^{(4)}_{\vec{k}\pm}(\Omega_{\vec{k}'}, \pm) = \frac{1}{2ik}(e^{2i\eta_0} - 1). \tag{21}$$

We neglect in this section the part p or f of the screening charge but we shall consider in Appendix C the result of a small additional phase shift η_1.

We proceed now to the calculation of the scattering intensity

$$|f_{k\pm}(\Omega_{\vec{k}'}, \pm)|^2 = |f^{(1)} + f^{(2)} + f^{(3s)} + f^{(3a)} + f^{(4)}|^2. \tag{22}$$

We have to select in this scattering intensity the terms which changes sign when $\vec{k}$ and $\vec{k}'$ are interchanged and which are of odd order in J_z and even order in s_z. Such terms arises obviously from the products of $f^{(2)}$ by $f^{(3s)}$ or $f^{(4)}$ and of $f^{(3a)}$ by $f^{(1)}$ (see also the discussion of Appendix B). When the antisymmetric terms of the scattering intensity are obtained, they have to be introduced in an expression of the skew-scattering Hall resistivity, for instance the expression (15) of Ref. 5 for elastic collisions:

$$\rho^{(\pm)}_{xy} = -\left(\frac{\hbar}{8\pi^3 ne}\right)^2 \int \left(-\frac{\partial f^0}{\partial \epsilon_{\vec{k}}}\right)(\vec{k}\cdot\vec{u})(\vec{k}'\cdot\vec{v}) \times w^a(\vec{k}\pm \rightarrow \vec{k}'\pm)\, d^3\vec{k}\, d^3\vec{k}', \tag{23}$$

where n is the number of electrons with a given spin direction, f^0 the Fermi-Dirac distribution, $\vec{u}$ and $\vec{v}$ are unit vectors in the directions x and y, and where $w^a(\vec{k}\pm \rightarrow \vec{k}'\pm)$ is the antisymmetric part of the scattering probability $w(\vec{k}\pm \rightarrow \vec{k}'\pm)$:

$$w(\vec{k}\pm \rightarrow \vec{k}'\pm) = \frac{8\pi^3\hbar^3}{m^2}|f_{\vec{k}\pm}(\Omega_{\vec{k}'}, \pm)|^2 \times \delta(\epsilon_{\vec{k}} - \epsilon_{\vec{k}'})\, n\, \mathrm{imp}. \tag{24}$$

The calculation of the integral can be simplified if one remarks that, on account of the factor $(\vec{k}\cdot\vec{u})(\vec{k}'\cdot\vec{v})$, the integral cancels out unless the dependence of $|f|^2$ on the directions of $\vec{k}$ and $\vec{k}'$ is that of p spherical harmonics ($l = 1$). This means that one must pick up in $|f|^2$ the products of two scattering amplitude terms which depend on $\vec{k}$ (or $\vec{k}'$) through spherical harmonics Y_l and $Y_{l'}$ with $(l + l')$ odd. So we have only to consider in $|f|^2$ the products of the antisymmetric term $f^{(2)}$ ($l = 1$) by $f^{(3s)}$ ($l' = 2$) or $f^{(4)}$ ($l' = 0$) and the products of the antisymmetric term $f^{(3a)}$ ($l = 2$) by the parts $l' = 1$ or $l' = 3$ of $f^{(1)}$. After integration one obtains

$$\rho^{(\pm)}_{xy} = -\frac{4\pi^2\hbar c N n(E_F)}{3ne^2k_F}\langle J_z\rangle\Big[(\sin^2\eta_0 - \sin^2\eta_2)(2-g)F_2 + \tfrac{9}{2}(\lambda/\Delta)\sin^2\eta_2 \sin 2\eta_2 \times (g-1)(\Gamma^{(1)} - \Gamma^{(3)})\Big]. \tag{25}$$

It can be noticed that the first term in the bracket provides a contribution proportional to $(2-g)$ $\langle J_z\rangle$, that is, to the orbital polarization of the RE, and the second a contribution proportional to $(g-1)\langle J_z\rangle$, that is to the spin polarization.

The total Hall resistivity is given by expression (13) of Ref. 5:

$$\rho_{xy} = \left[1 + \left(\frac{i^+ - i^-}{i^+ + i^-}\right)^2\right]\left[R_0 H + \tfrac{1}{4}(\rho^+_{xy} + \rho^-_{xy})\right] + \left(\frac{i^+ - i^-}{i^+ + i^-}\right)\left(\frac{\rho^+_{xy} - \rho^-_{xy}}{2}\right) \tag{26}$$

(R_0 is the coefficient of ordinary Hall effect, i^+ and i^- are the currents for each spin direction; note the correction of a factor $\frac{1}{2}$ in Ref. 5 into $\frac{1}{4}$). For rare-earth impurities in Ag, Au, or Al the spin-independent scattering is most dominant,[9] i^+ and i^- are nearly equal, and (26) becomes

$$\rho_{xy} = R_0 H + \tfrac{1}{4}(\rho^+_{xy} + \rho^-_{xy}) \tag{27}$$

(we shall consider in Appendix C the additional terms resulting from the occurrence of slightly different values for i^+ and i^-).

In the temperature range where the magnetic susceptibility reaches the free-ion susceptibility nearly (kT greater than the crystal-field splitting) and at low field $\langle J_z\rangle$ can be written

$$\langle J_z\rangle = -gJ(J+1)\mu_B H/3k_B T \quad (g\mu_B = |g\mu_B|). \tag{28}$$

We also introduce the impurity resistivity ρ_0, which is mostly due to the spin-independent phase shifts η_0 and η_2, and can be written

$$\rho_0 = (2\pi\hbar C N/ne^2k_F)(\sin^2\eta_0 + 5\sin^2\eta_2). \tag{29}$$

Then, from (25), (27), and (28), we can write the Hall coefficient ($R = \rho_{xy}/H$) in a form similar to (4):

$$R = R_0 + (a_1 + a_2)\rho_0 T^{-1}, \tag{30}$$

with

$$a_1 = \frac{\pi\mu_B}{9k_B}\,\frac{\sin^2\eta_0 - \sin^2\eta_2}{\sin^2\eta_0 + 5\sin^2\eta_2}$$

$$\times n(E_F)F_2 g(2-g)J(J+1), \qquad (31)$$

$$a_2 = \frac{\pi\mu_B\lambda}{2k_B\Delta}\,\frac{\sin^2\eta_2 \sin 2\eta_2}{\sin^2\eta_0 + 5\sin^2\eta_2}$$

$$\times n(E_F)(\Gamma^{(1)} - \Gamma^{(3)})g(g-1)J(J+1). \qquad (32)$$

In a given host, η_0, η_2, λ, Δ, F_2, $\Gamma^{(3)}$, and $\Gamma^{(1)}$ may be supposed nearly independent of the RE element. Then a_1 should vary throughout the RE series like the factor $g(2-g)J(J+1)$ and a_2 like $g(g-1)J(J+1)$.

V. DISCUSSION

The analysis of the crystal-field effects on the skew scattering at low temperature is not in the scope of this paper, and we shall only discuss the experimental results in the temperature range where the magnetic susceptibility[17,20] reaches nearly its free-ion value. According to Sec. III, the experimental Hall coefficient in this temperature range can be written [expression (4)]

$$R = R_0 + a\rho_0 T^{-1}.$$

The coefficient a is characteristic of the skew scattering by a given impurity in a given host. The experimental values of a for the Ag-, Au-, and Al-based alloys are plotted throughout the RE series on the Figs. 12, 13, and 15.

The calculation of Sec. IV predicts that a is the sum of a term a_1 varying in the RE series like $g(2-g)J(J+1)$ and of a term a_2 varying like $g(g-1)J(J+1)$. We tried to fit the experimental variation of a throughout the RE series with a linear combination of $g(2-g)J(J+1)$ and $g(g-1)J(J+1)$.

For the Ag:*R* alloys the best fit is obtained with

$$a = 0.43\times 10^{-8} g(2-g)J(J+1)$$

$$-1.57\times 10^{-8} g(g-1)J(J+1) \quad (a \text{ in K/G}). \qquad (33)$$

It can be seen on Fig. 12 that the variation of the calculated values in the RE series reproduces fairly well the experimental one. The competition between the terms a_1 and a_2 explains that a, maximum for Gd, decreases more rapidly than $g(g-1)J(J+1)$ for Tb, Dy, etc., ... and changes sign for Er. For the light RE a_1 and a_2 are both positive, what is consistent with the positive experimental values of a. However the quantitative agreement is less good for the light RE than for the heavy.

For the Au:*R* alloys, the best fit is obtained with

$$a = 0.34\times 10^{-8} g(2-g)J(J+1)$$

$$-2.26\times 10^{-8} g(g-1)J(J+1) \quad (a \text{ in K/G}). \qquad (34)$$

A term a_2 larger than in the Ag:*R* alloys explains that a is larger in gold and does not change sign in the heavy RE series. For the light RE, the agreement is satisfying. For *Au*Yb we estimate $a = 2\times 10^{-8}$ K/G from the low-temperature data after correction of the CEF effects. This value is larger than the calculated one [from (34)] and this is certainly due to the "anomalous" character of the Yb impurities (admixture between conduction and f electrons). So the case of the *Au*Yb alloy is relevant to another theory of skew scattering.[5]

For the Al:*R* alloys, the experimental values of a cannot be treated with the same confidence than that for the Ag:*R* or Au:*R* alloys on account of the doubt on the metallurgy. The fit shown on Fig. 15 corresponds to

$$a = 0.32\times 10^{-8} g(2-g)J(J+1)$$

$$-0.63\times 10^{-8} g(g-1)J(J+1) \quad (a \text{ in K/G}). \qquad (35)$$

The fit of the experimental results with combinations of a_1 and a_2 provides interesting information on the antisymmetric terms of the electron-impurity interaction.

By identifying the first term of (33) to (31) one obtains for the Ag:*R* alloys

$$\frac{\pi\mu_B}{9k}\,\frac{\sin^2\eta_0 - \sin^2\eta_2}{\sin^2\eta_0 + 5\sin^2\eta_2}\, n(E_F)F_2 = 0.43\times 10^{-8}\ \text{K/G}.$$

We take as screening charge $Z_s = 1$, $Z_p = 0$, $Z_d = 1$, which gives $\eta_0 = \frac{1}{2}\pi$, $\eta_2 = \frac{1}{10}\pi$, and a residual resistivity $\rho_0 = 6.42$ μΩcm/at.%, in agreement with the experimental residual resistivities (from 5.3μΩcm/at.% for Gd to 6.7 for Dy in the heavy-RE series). With $\eta(E_F) = 0.15$ states/eV atom spin, one obtains

$$F_2 = 2\times 10^{-3}\ \text{eV}.$$

The choice of Z_s and Z_d is not very crucial. If one lets Z_d free to vary from 0.5 to 1.5 (always with $Z_s + Z_d = 2$), F_2 varies from 1.6×10^{-3}eV to 6.5×10^{-3} eV.

The same calculation for the Au:*R* alloys, with $Z_d = +1$, $Z_s = +1$, provides

$$F_2 = 1.6\times 10^{-3}\ \text{eV}.$$

In the Al:*R* alloys, F_2 appears to be of the same order of magnitude.

Thus we find that the coupling between the orbital angular momentum of the conduction electrons and the total angular momentum of the RE is rather small, about 50 times smaller than the spin-spin exchange coupling $\Gamma^{(0)}$. It turns out, for example, that the contribution of such a coupling to the relaxation of RE impurities in EPR—proportional to the square of F_2—should be negligible. The contribution of the $\vec{1}\cdot\vec{J}$ coupling to the RE g shift through the orbital polarization of the conduction electrons ($\Delta g \sim F_2$) should also be small, but perhaps nonnegligible.

By identifying the second terms of (33) to (32), one obtains for the Ag:R alloys

$$\frac{\pi\mu_B}{2k}\frac{\lambda}{\Delta}\frac{\sin^2\eta_2 \sin 2\eta_2}{\sin^2\eta_0 + 5\sin^2\eta_2}\eta(E_F)(\Gamma^{(1)} - \Gamma^{(3)}) = -1.5\times 10^{-8}\ \mathrm{K/G}. \tag{36}$$

This relation could allow us to deterime $\Gamma^{(1)} - \Gamma^{(3)}$. However this determination is much less direct than that of F_2 as the expression (36) contains, in addition to $\sin\eta_0$ and $\sin\eta_2$, the parameters λ and Δ which are only known approximately. Callender and Schnatterly[29] have deduced from the optical properties of *Ag*Pd alloys a half-width of the $4d$ vbs of Pd equal to 0.25 eV. Taking the same value of Δ for the $5d$ vbs of the RE, $\lambda = 0.1$ eV, $\eta_0 = \frac{1}{2}\pi\,(Z_s = +1)$ and $\eta_2 = \frac{1}{10}\pi\,(Z_d = +1)$, we obtain

$$\Gamma^{(3)} - \Gamma^{(1)} = 0.06\ \mathrm{eV}.$$

This seems somewhat too large as, after Kondo,[14] one expects the same order of magnitude for F_2 and $\Gamma^{(1)}$ or $\Gamma^{(3)}$ ($\Gamma^{(1)}$ corresponds to the coefficient F_1 in the paper of Kondo). So we present in Appendix C an alternative interpretation of the magnitude of a_2 by means of an additional skew-scattering term which results from the difference between the spin-up and spin-down currents and which is also proportional to $(g-1)\langle J_z\rangle$. We emphasize that, on account of the several terms involved, the interpretation of the magnitude of a_2 is complex and does not really allow us a determination of $\Gamma^{(1)}$, $\Gamma^{(3)}$, or Δ.

For the Au:R alloys, the term a_2 is larger than for the Ag:R, what could be explained by a narrower vbs. With the same value of $\Gamma^{(3)} - \Gamma^{(1)}$, one obtains $\Delta_{\mathrm{Au}} = 0.7\Delta_{\mathrm{Ag}} = 0.175$ eV.

For the Al:R, the term a_2 is much smaller. It could correspond to a much wider $5d$ vbs ($\Delta \simeq 1$ eV). These different values of Δ in Au, Ag, and Al are not surprising as it is well known that transition-metal impurities are less magnetic in Al than in Ag, and less magnetic in Ag than in Au.

VI. SUMMARY AND CONCLUSIONS

This paper report a systematic study of the skew scattering by rare-earth impurities in silver, gold, and aluminium. The main features of the Hall effect induced by skew scattering are collected in Figs. 1–11. We have not analyzed the crystal-field effects at low temperature, and we have concentrated on the results at "high temperature." We have proposed a model which explains, in particular, the variation of the skew scattering in the RE series.

The most interesting information concerns the term $\vec{1}\cdot\vec{J}$ of exchange coupling between the orbital angular momentum of the conductions electrons and that of the $4f$ electrons. It turns out that this coupling is smaller than the spin-spin exchange coupling by almost two orders of magnitude. It is interesting to compare this result to that of resistivity anisotropy measurements[10] on the same alloys which show that another type of anisotropic interaction, the Coulomb interaction between a conduction electron and the electric quadrupole of a RE ion, is much stronger, sometimes as strong as the spin-spin exchange interaction.

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APPENDIX A: GENERALITIES ON SKEW SCATTERING

Skew scattering arises when the scattering potential includes an antisymmetric part H^A:

$$\langle \vec{k}'|H^A|\vec{k}\rangle = -\langle \vec{k}|H^A|\vec{k}'\rangle = \text{imaginary}.$$

Let us assume very generally a scattering potential

$$H^S = \sum_{\vec{k}\vec{k}'} a^*_{\vec{k}'} a_{\vec{k}} (V_{\vec{k}\vec{k}'} + iW_{\vec{k}\vec{k}'}),$$

where $V_{\vec{k}\vec{k}'}$ is real and symmetric, $W_{\vec{k}\vec{k}'}$ real and antisymmetric. In the first Born approximation, the T matrix elements are

$$T^{(1)}_{\vec{k}\vec{k}'} = V_{\vec{k}\vec{k}'} + i\,W_{\vec{k}\vec{k}'}, \tag{37}$$

and the scattering probability is symmetric (nonskew):

$$W^{(1)}(\vec{k}\to\vec{k}') = (2\pi/\hbar)|T^{(1)}_{\vec{k}\vec{k}'}|^2 = (2\pi/\hbar)(V^2_{\vec{k}\vec{k}'} + W^2_{\vec{k}\vec{k}'}).$$

Antisymmetric terms appear only if the T matrix is calculated to second Born approximation; the second order of $T_{\vec{k}\vec{k}'}$ includes terms such as

$$\sum_{\vec{k}''} \frac{V_{\vec{k}\vec{k}''} V_{\vec{k}\vec{k}''}}{\epsilon_k - \epsilon_{k''} + is} = -i\pi \sum_{\vec{k}''} V_{\vec{k}\vec{k}''} V_{\vec{k}''\vec{k}}\,\delta(\epsilon_k - \epsilon_{k''}) + \text{real term}. \tag{38}$$

It turns out that this imaginary symmetric term can interfere with the imaginary antisymmetric term $iW_{\vec{k}\vec{k}'}$ in $T^{(1)}_{\vec{k}\vec{k}'}$. The scattering probability then includes antisymmetric terms such as

$$-\frac{2\pi^2}{\hbar}\sum_{k''} W_{\vec{k}\vec{k}''} V_{\vec{k}\vec{k}'} V_{\vec{k}''\vec{k}'}\,\delta(\epsilon_k - \epsilon_{k''}).$$

So the lowest skew terms of the scattering probability are of first order in W and second order in V. There are also terms third order in W but they will be generally much smaller as $W \ll V$.

APPENDIX B: SELECTION OF THE SKEW-SCATTERING TERMS

We consider the scattering by a RE impurity with the following scattering potential:

$$H = -(g-1)\Gamma\delta(r)\vec{s}\cdot\vec{J} - (2-g)\Lambda\vec{l}\circ\vec{J}$$
$$+ V\delta(r) + \lambda'\vec{l}\cdot\vec{s},$$

where the two first terms correspont to $H^{(1)}$ and $H^{(2)}$ of Sec. IV and the two last term to $H^{(3)}$[with $\Lambda \sim F_2$, $\lambda' \sim \lambda\Delta^{-1}n(E_F)^{-1}\sin^2\eta_2$]. Actually these terms have in Sec. IV a more complicated spatial dependence. But the simplification to $\Gamma\delta(r)$ or $V\delta(r)$ does not matter here as we want only to select the skew terms in the scattering probability without calculating them.

The skew terms of the transition probability correspond, according to Appendix A, to products of one antisymmetric term of H by two symmetric terms. We show first that these triple products can only involve the non-spin-flip part of H. Suppose that the triple product includes antisymmetric terms $l^{\pm}J^{\mp}$ or $l^{\pm}s^{\mp}$ which flip either $\vec{J}$ or $\vec{s}$. One cannot come back to the initial state with $s^{\pm}J^{\mp}$ which flips $\vec{s}$ and $\vec{J}$ at the same time. However there are coherent triple products including twice s^+J^- (or s^-J^+) or both s^+J^- or s^-J^+. But these products are of order Γ^2 and so much smaller than the triple products of order V^2 or $V\Gamma$ as $V \gg \Gamma$. So H can be limited to its non-spin-flip part H':

$$H' = -(g-1)\Gamma\delta(r)J_zS_z - (2-g)\Lambda l_zJ_z$$
$$+ V\delta(r) + \lambda' l_zs_z\,.$$

With the antisymmetric term $(2-g)\Lambda l_zJ_z$ one obtains triple products which are, respectively, proportional to $(2-g)\Lambda V^2J_z$, $(2-g)\Lambda V(g-1)\Gamma s_zJ_z^2$, and $(2-g)\Lambda(g-1)^2\Gamma^2s_z^2J_z^3$. As $V \gg \Gamma$, one keeps only the first product. It correspond to the first Hall-resistivity term in (25). As this term is of first order in Λ, it is sufficient to study the scattering of the p waves by $H^{(2)}$ in first order. This justifies the calculation of $f^{(2)}$ in first order.

With the antisymmetric term $\lambda' l_zs_z$ of H' one obtains triple products, respectively, proportional to $\lambda' V^2s_z$, $\lambda' V(g-1)\Gamma s_z^2J_z$, and $\lambda'(g-1)^2\Gamma^2s_z^3J_z^2$. The only term odd in J_z and even in s_z is the second. It corresponds to the second Hall-resistivity term in (25). As this term is of first order in Γ, it is sufficient to study the scattering of the p and f waves in first order in $H^{(1)}$. This justifies the calculation of $f^{(1)}$ in Sec. IV.

The same sort of considerations shows that the third term in (2) contributes to the Hall resistivity with a term proportional to $F_3V\Gamma\langle s_z^2\rangle\langle J_z\rangle$, which is certainly small if $\Gamma \ll V$.

Thus simple considerations of symmetry and order of magnitude allows us to select the main skew terms and justifies the calculation of Sec. IV.

APPENDIX C: HALL-RESISTIVITY CALCULATION WITH DIFFERENT SPIN-UP AND SPIN-DOWN CURRENTS

For rare-earth impurities in silver, gold, or aluminium, the spin-dependent scattering is dominant and so, in the calculation of Sec. IV, we have supposed equal spin-up and spin-down currents ($i^+ = i^-$). We shall now take into account the small difference between i^+ and i^- induced by the exchange interaction when the impurities are polarized by a magnetic field; we shall also suppose that there is a part p in the screening charge in addition to the parts s and d, and we shall calculate the resulting additional terms of Hall resistivity.

Considering the scattering amplitude of Sec. IV plus a new term accounting for the phase shift η_1, we calculate first the difference of scattering rates for the spin-up and spin-down electrons and the difference between the currents i^+ and i^-.

The calculation is similar to the classical one of Ref. 30 and leads to

$$r = \frac{i^+ - i^-}{i^+ + i^-} = -\pi\,\frac{\sin(\eta_0-\eta_1)\cos(\eta_0-\eta_1)}{\sin^2(\eta_0-\eta_1) + 2\sin^2(\eta_1-\eta_2) + 3\sin^2\eta_2}\,(g-1)\Gamma^{(0)}n(E_F)\langle J_z\rangle \tag{39}$$

(we have limited the expression of r to terms in $\Gamma^{(0)}$).

For rare-earth impurities in Ag, Au, or Al, r is much smaller than one and we shall use the only Hall-resistivity term of first order in r, that is, the last term in expression (26). It provide an additional contribution to the Hall resistivity:

$$\rho'_{xy} = \frac{i^+ - i^-}{i^+ + i^-} \times \tfrac{1}{2}(\rho^+_{xy} - \rho^-_{xy}). \tag{40}$$

In Sec. IV we have calculated the only terms ρ^+_{xy} and ρ^-_{xy} which are odd in J_z and even in s_z. Calculating also the terms odd in s_z and selecting the most important by considerations of order of magnitude (as in Appendix B) we obtain

$$\rho^+_{xy} - \rho^-_{xy} = -[24\pi^2\hbar c N n(E_F)/ne^2k_F] \times (\lambda/\Delta)\sin^2\eta_2 \sin(2\eta_2 - \eta_1)\sin\eta_1. \tag{41}$$

By introducing (39) and (41) in (40) we obtain a new Hall-resistivity term proportional to $(g-1)\Gamma^{(0)}\langle J_z\rangle$, and so an additional contribution to a_2, obtained by replacing $(\Gamma^{(1)} - \Gamma^{(3)})\sin^2\eta_2$ in (32) by

$$-4\Gamma^{(0)} \frac{\sin(\eta_0 - \eta_1)\cos(\eta_0 - \eta_1)\sin(2\eta_1 - \eta_1)\sin\eta_1}{\sin^2(\eta_0 - \eta_1) + 2\sin^2(\eta_1 - \eta_2) + 3\sin^2\eta_2}. \tag{42}$$

Taking the same values of λ, Δ, $n(E_F)$ as in Sec. IV and the phase shifts which are derived from the interpretation of the resistivity and of the negative magnetoresistance of[10] *Au*Gd ($\eta_0 = 113.4°$ or $Z_s = 1.26$, $\eta_1 = -9°$ or $Z_p = -0.3$, $\eta_2 = 18.7°$ or $Z_d = 1.04$ with $Z_s + Z_p + Z_d = 2$), one calculates a value of a_2 which is smaller by only 15% than the experimental value for the Ag:*R* alloys. So it appears that the magnitude of a_2 can be explained in this way, without invoking a large value of $(\Gamma^{(3)} - \Gamma^{(1)})$ as in Sec. IV.

There is also a term involving r^2 in expression (26) of Hall resistivity. From expression (39) and also from the interpretation of the negative magnetoresistance[10] it is possible to show that r^2 remains smaller than about 10^{-3} for Ag:*R* or Au:*R* and that the contribution of the term in r^2, in particular, the "spin effect" on the ordinary Hall effect, is negligible certainly (anyhow the term in r^2 is proportional to H^3 at low field and so does not contribute to the initial Hall coefficient).

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