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# Hall effect in amorphous calcium-aluminum alloys

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We present results of the Hall effect measurements in CaAl(Au) amorphous alloys. The Hall coefficients have been found to be negative and independent of temperature. Their magnitudes deviate significantly from the nearly-free-electron calculations, and are reduced by gold doping. These deviations have been accounted for from considerations of the unusual electronic structure of CaAl, and the effects of both $s$-$d$ hybridization and side-jump mechanism on the conduction electrons.

## I. INTRODUCTION

The Hall effect is an important electronic transport property which results from the effect of the Lorentz force acting on the conduction electrons. In simple liquid and amorphous metals, the Hall coefficient is negative with a magnitude close to the free-electron value. The Hall coefficient of amorphous alloys containing transition metals is often positive, contrary to the free-electron expectation.[1,2] The observation of a positive Hall coefficient gave rise to intense interest in the study of the Hall effect in amorphous alloys and a number of different theoretical models have been proposed to explain this anomalous behavior. Using the nearly-free-electron model, the Hall coefficient is expected to be negative in materials with a spherically symmetric Fermi surface, unless the group velocity at the Fermi surface is negative.[3,4] At present, there is a controversy about the origin of the positive Hall coefficient in these materials. Weir *et al.*[5] calculated the effect of $s$-$d$ hybridization on the free-electron band in amorphous alloys and obtained an $S$-shaped dispersion relation $E(k)$ for the $s$ band in the region of the $d$ band. The electron-group velocity, defined from the slope of the dispersion relation, changes sign from positive to negative in this region and leads to the occurrence of a positive Hall coefficient if the Fermi level lies in this region. A model for the Hall coefficient in amorphous alloys based on the theory of $s$-$d$ hybridization using linear-response theory, was developed by Howson and Morgan[6] to account for this observation.

An alternative explanation of the positive Hall coefficient is based on the side-jump mechanism originally proposed by Berger[7] to explain the anomalous Hall effect in ferromagnetic alloys. Recently Stobiecki and Przybylski[8] and Trudeau *et al.*[9,10] have suggested that the side-jump mechanism might also explain the positive Hall effect in amorphous alloys. In particular, they were able to explain the strong temperature dependence of the Hall effect in Fe-Zr and Y-Al alloys, which correlated with the magnetic susceptibility. The side-jump (SJ) mechanism results from the interaction of the conduction-electron spin and the atomic-orbital moment of the scattering center and gives an additional term to the normal Hall coefficient. This additional term can give rise to a positive Hall coefficient if it is positive and sufficiently large to overcome the negative contribution of the normal term. Rhie and Naugle[11] used the same effect to explain the composition dependence of the Hall coefficient in Zr-Ni alloys. It is important to note that, while the SJ mechanism leads to an extra contribution to the Hall effect, $s$-$d$ hybridization changes the sign of the normal Lorentz force contribution itself. In principle, both effects can be present.

There has been special interest in the study of the Hall effect in CaAl amorphous alloys, since these alloys are composed of simple metals but have large values of electrical resistivities, ranging from 140 to 500 $\mu\Omega$ cm and exhibit anomalous transport properties. The presence of Ca $d$ states at the Fermi level[12,13] and the hybridization between the $s$ and $d$ states may significantly alter the form of the Hall coefficient in comparison to that expected from the free-electron model. The large values of the electrical resistivities result from a combination of strong $s$-$d$ scattering and quantum interference effects.[14] These large values of electrical resistivities and the strong spin-orbit scattering from gold doping are used to test whether the side-jump hypothesis can produce a positive contribution to the Hall effect in CaAl amorphous alloys.

In this study, the effect of increasing the strength of spin-orbit scattering by gold doping on the Hall coefficient has been investigated and an attempt has been made to account for the differences between the measured Hall coefficients and the calculated free-electron values within the framework of both $s$-$d$ hybridization and side-jump mechanism models.

## II. EXPERIMENTAL TECHNIQUES

The measurements of the Hall coefficients were made in the temperature range 1.5–25 K, and in magnetic fields up to 7 T. The samples were prepared by radio-frequency sputtering at a pressure of $10^{-3}$ mbar of Ar. During the sputtering process, the substrate holder was oscillated back and forth over the target to ensure a homogeneous film deposition. The target was made from rolled aluminum and cut into a disk of diameter 50 mm. Segments of calcium metal and short pieces of thin gold wire (for gold-doped samples) were placed on top of the aluminum disk. Samples of different compositions were obtained by varying the number of calcium and gold

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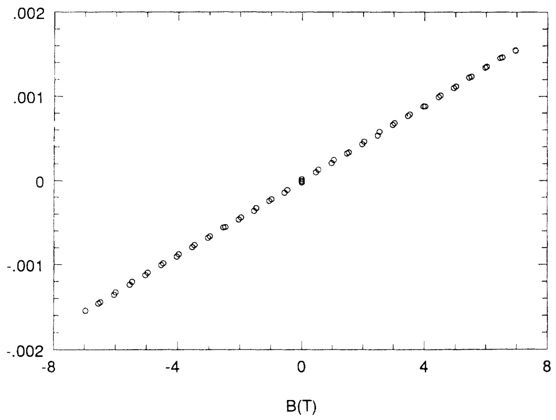

FIG. 1. Hall resistance of $Ca_{45}Al_{55}$ measured in both forward and reverse field directions at $T=5.0$ K.

pieces. The substrates used were glass slides of dimensions $18.2\times12.1\times1.0$ mm. Prior to sputtering, the glass slides were immersed in a solution of dilute nitric acid (20%) and left overnight to degrease and then cleaned for about 30 min using energetic argon atoms from a fast atom source in the sputtering chamber. Platinum contacts were initially sputtered onto the glass slides, which provided suitable areas to which we could solder. The sample was deposited through a two-probe crucifixial contact mask aligned with the platinum pads. This method produced misalignment voltage of the order of 10 $\mu$V. Before depositing a sample onto the substrate, a shutter was placed above the target for about 15 min in order to avoid unwanted oxides on the surface of the target from depositing on the substrate. Films of typical thickness ~1 $\mu$m were obtained.

The chemical composition of the sputtered thin films was obtained by electron microprobe analysis which has a detection limit of 200 ppm. This was carried out on films deposited on small pieces of copper plates which were mounted close to the glass slides, during the sputtering process. The thickness of the sputtered samples were measured using a Sloan Dectak Profilometer which uses a stylus to give thickness profiles directly. The Hall resistances were measured using a Keithley 181 DVM with a precision of 20 $\mu\Omega$. In order to correct for the magnetoresistance arising from the misalignment of the Hall probes, the Hall resistances were measured in both forward and reverse field directions as shown in Fig. 1 for the $Ca_{45}Al_{55}$ sample. All samples showed similar linear dependence of the Hall resistance as a function of the magnetic field. The Hall coefficient is calculated from the product of the sample thickness and the slope obtained from the least-squares fit of the plot of the Hall resistance against the magnetic field. The sign of the Hall coefficient was checked by comparison with that of a copper sample.

## III. THEORETICAL REVIEW

The development of the theory of the Hall effect in amorphous alloys has evolved from that applied to liquid metals. This is mainly because of their structural similarity. Both liquid and simple metal amorphous alloys are isotropic and are expected to have a nearly-free-electron-like spherical Fermi surface (broadened by the disorder). The expression of the Hall coefficient using a semiclassical approach is given by[3]

$$R_H=\frac{\alpha}{ne}\,, \tag{1}$$

where $n$ is the number of conduction electrons per unit volume, $e$ is the electronic charge, and $\alpha$ is close to unity. In this model, the Hall coefficient depends only on the electron density and takes the free-electron value in both liquid and simple metal amorphous alloys with a sign opposite to that of the group velocity.

Significant deviations from this description are observed in high-resistivity amorphous alloys which are usually based on transition metals and their Hall coefficients are often positive.[1,15,16,17] In these materials, the basic assumptions of the nearly-free-electron model become inadequate because of the short electron mean free paths. There have been a number of approaches to account for these deviations from the free-electron behavior, the most significant of which are the side-jump mechanism and the $s$-$d$ hybridization.

### A. The side-jump mechanism

The side-jump mechanism refers to an effective transverse displacement of the conduction electron due to spin-orbit scattering. The magnitude of the transverse displacement depends on the strength of the spin-orbit scattering and it is argued that for $d$ electrons, the transverse displacement is large enough to give a significant contribution to the Hall effect.[10,18]

The Hall coefficient, according to this hypothesis, is composed of two terms:[9]

$$R_H=R_0+R_s\chi\,, \tag{2}$$

where $R_0$ is the normal Hall coefficient from the effect of the Lorentz force on the charge carriers and $R_s\chi$ is the contribution from the side-jump effect. The parameter $\chi$ is the electron susceptibility and $R_s$ is given by[9]

$$R_s=\frac{2e^2\lambda_{so}\rho^2}{\hbar\mu_\sigma\mu_B g^*}\,. \tag{3}$$

In this expression, $\rho$ denotes the electrical resistivity and $\lambda_{so}$ is the effective spin-orbit coupling parameter. Other symbols have their usual meaning. If the value of $R_s\chi$ is large and positive relative to $R_0$, then the resultant Hall coefficient would be positive. The sign of $R_s$ depends on the sign of $\lambda_{so}$ which, in turn, depends on the position of the Fermi level $E_F$ in the $d$ band. When the Fermi level passes through the middle of the band, $\lambda_{so}$ changes sign and it is, thus, possible on this basis to explain the dependence of the sign of the Hall coefficient on the composition of the alloy.[11,19]

### B. *s*-*d* hybridization

When a free-electron-like $s$ band overlaps with the $d$ band, $s$-$d$ hybridization results in changes in the $s$ band's electron dynamics and can lead to a negative group velocity for the hybridized $s$ band.[5,6] Since the sign of the Hall coefficient depends on the sign of the electron-group velocity, $V=\hbar^{-1}dE/dk$,[3] it is then possible to obtain a positive Hall coefficient from the dispersion relation $E(k)$ when $dE/dk$ is negative. This has been shown in calculations by Morgan, Howson, and Nguyen-Manh[20] and Weir *et al.*[5] In their approach, the scattering of plane-wave states into other free-electron-like states via a weak pseudopotential is ignored, so that only the scattering of the plane-wave states into $d$ states is considered. The shift in energy of the free-electron states hybridized with $d$ states is obtained by evaluating the self-energy $\gamma(k,E)$ corresponding to the diagonal Green's function $G=(E-k^2-\gamma)^{-1}$. An $S$-shaped dispersion relation $[E(k)]$ is obtained by using a suitable model of the "$d$"—density of states and appropriate parameters for the $s$-$d$ coupling.[5] When the Fermi level lies in the region in which $dE/dk$ is negative, the sign of the Hall coefficient is positive. Thus, the $S$-shaped curve of the dispersion relation for the hybridized $s$ band provides a possible explanation for the existence of the positive Hall coefficient in amorphous alloys. The problem with this model is understanding the significance of the group velocity when the lifetime is small in the region of anomalous dispersion. However, detailed calculations[5,6] show that despite significant broadening of the dispersion curve by disorder scattering, the arguments in terms of the sign of $dE/dk$ work well.

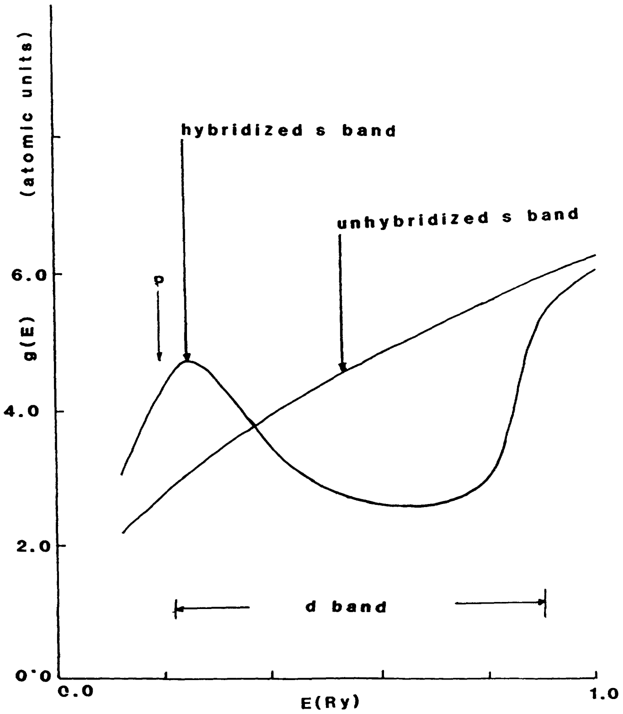

FIG. 2. Effect of $s$-$d$ hybridization on the density of $s$ states Ref. 5. The region of the $d$ band is indicated.

An alternative approach based on $s$-$d$ hybridization involves an approximate calculation of the Hall coefficient from the derivative of the density of states in the weak scattering limit:[21]

$$R_H=-\frac{\alpha g'(E_F)}{2|e|cg^2(E_F)}\,, \tag{4}$$

where $g'(E_F)$ is the derivative of the "$s$" density of states at the Fermi level, $\alpha=3$ for free electrons, and $\alpha\sim 1$ in general. Hybridization between the $s$ and $d$ bands reduces the density of the free-electron states in the region of the $d$ band as shown in Fig. 2. Thus, the sign of the Hall coefficient using Eq. (4) depends on the sign of the derivative of the density of states, $g'(E_F)$ at the Fermi level. This approach is related to that using the anomalous dispersion relation, since $s$-$d$ hybridization produces both an $S$-shaped dispersion relation and a minimum in the density of states.[22,23] It is important to note that when $E_F$ is within the $d$ band, $g'$ can be negative and the Hall coefficient positive. But when $E_F$ is outside the region of the $d$ band, the modification of $g$ and $g'$ can lead to a negative Hall effect which is significantly enhanced over the free-electron value.

## IV. RESULTS AND DISCUSSION

Table I gives the values of the Hall coefficients and electrical resistivities of $(\mathrm{Ca}_x\mathrm{Al}_{100-x})_{100-y}\mathrm{Au}_y$. It is also instructive to estimate values for the average number of conduction electrons per atom $n_A(\equiv e/a)$ and $k_Fl$, where $k_F$ is the Fermi wave vector and $l$ the electron mean free path. These values have been obtained from the measured Hall coefficients and electrical resistivities by applying the free-electron expressions. The values of $k_Fl$ are consistent with strong scattering systems in which the mean free path is of the order of a few atomic diameters and $k_Fl$ is close to unity, except for $(\mathrm{Ca}_{81}\mathrm{Al}_{19})_{99.0}\mathrm{Au}_{1.0}$ with the lowest electrical resistivity of the series. The resistivity for $\mathrm{Ca}_x\mathrm{Al}_{100-x}$ has been shown to rise sharply from $\sim 140\ \mu\Omega\,\mathrm{cm}$ for $x=80$ to $\sim 500\ \mu\Omega\,\mathrm{cm}$ for $x\simeq 60$.[24,25]

TABLE I. Hall-effect data for $(\mathrm{Ca}_x\mathrm{Al}_{100-x})_{100-y}\mathrm{Au}_y$.

| Alloy | $\rho$ ($\mu\Omega\,\mathrm{cm}$) | $-R_H$ ($10^{-11}\ \mathrm{m^3C^{-1}}$) | $e/a$ | $k_Fl$ |
|---|---|---|---|---|
| $y=0.0, x=45$ | 384 | 6.3 | 2.5 | 2.2 |
| $x=47$ | 350 | 9.7 | 1.6 | 2.8 |
| $x=49$ | 327 | 10.4 | 1.6 | 3.1 |
| $x=64$ | 347 | 17.5 | 1.1 | 3.4 |
| $y=1.0, x=56$ | 426 | 9.5 | 1.8 | 2.3 |
| $x=61$ | 356 | 10.4 | 1.7 | 2.8 |
| $x=67$ | 379 | 14.8 | 1.5 | 3.0 |
| $x=81$ | 133 | 15.0 | 1.4 | 8.5 |
| $y=2.0, x=49$ | 381 | 8.4 | 1.8 | 2.4 |
| $x=52$ | 394 | 9.8 | 1.6 | 2.5 |
| $x=54$ | 469 | 11.0 | 1.4 | 2.2 |

Figure 3 shows the Hall coefficient of $Ca_xAl_{100-x}$, including the data of Mizutani and Matsuda[26] for comparison. The figure also includes the values of the Hall coefficient calculated from the nearly-free-electron model assuming two electrons per atom for Ca and three electrons per atom for Al.

The measured Hall coefficients of CaAl amorphous alloys are negative but are significantly larger in magnitude compared with the calculated free-electron values. The difference in the measured and calculated free-electron values of the Hall coefficient can be attributed to the unusual electronic structure of CaAl, with the presence of Ca *d* states at the Fermi level. Increasing the concentration of Al in CaAl pushes the Fermi level $E_F$ into the tail of the *d* band, and coupling between the *s* and *d* states then becomes possible at $E_F$. However, the number of *d* states is not sufficient to give a significant contribution to the conductivity[27] but the free-electron-like states are strongly scattered into the *d* states. The scattering of *s* states into the *d* states gives rise to a rapid increase in the electrical resistivity and *s*-*d* hybridization can significantly modify the normal Hall effect. Qualitatively, we can argue that in CaAl, the Fermi level $E_F$ lies in the vicinity of the point labeled *P* (refer to Fig. 2) in the tail of the Ca *d* states. In this region, $g'(E_F)$ is positive so the Hall effect is negative but the magnitude of the Hall coefficient is affected by the *s*-*d* hybridization. Both $g(E_F)$ and $g'(E_F)$ are larger than the free-electron values but the overall effect leads to an increase in $|R_H|$ over the free-electron value. The rapid drop in the Hall coefficient with the increase in the Al concentration as shown in Fig. 3, can be attributed to the approach of $E_F$ to the peak in the density of states in Fig. 2. Figure 4 shows the effects of gold doping on the Hall coefficient. It is clear from the figure that there is a significant reduction in the magnitude of the Hall coefficient by gold doping. The effect of increasing the Al concentration which has three electrons per atom compared with Ca which has two electrons per atom is to decrease the magnitude of the Hall effect as the Fermi Energy is pushed toward the peak in Fig. 2. The effect of Au, which contributes only one electron per atom, is, therefore, similar to removing Al. This being the case, we can estimate, using results in Fig. 3, that the addition of 1% Au to CaAl would actually *increase* the magnitude of the Hall coefficient by about 10%. But the measured Hall coefficient *decreases* with the addition of gold. A possible cause of the *decrease* in the measured values could be the side-jump effect. The effective spin-orbit interaction parameter $\lambda_{so}$ in CaAl is positive since $E_F$ lies at the lower edge of the *d* band and, therefore, the side-jump term, $R_s\chi$ can be positive. It is difficult to calculate the side-jump contribution. However, in order to estimate the magnitude of $R_s\chi$ in CaAl(Au), we deduced the values of $\lambda_{so}$ and $R_s$ (refer to Table II) from the values of resistivity $\rho$ and spin-orbit scattering rate $\tau_{so}^{-1}$ in YAl and CaAl(Au) (Refs. 10,28) assuming $\lambda_{so}\alpha\tau_{so}^{-1}$. Using the value of the electronic susceptibility, $\chi \simeq 2.5\times10^{-4}JT^{-2}$ $mol^{-1}$ for CaAl (Ref. 26) and the measured resistivity (140–400 $\mu\Omega$ cm) we estimate the additional side-jump term $R_s\chi$ to be of the order of between $5\times10^{-11}$ and $4\times10^{-10}$ $m^3C^{-1}$. These values are a little larger in magnitude than the observed drop in $R_H$ on addition of Au. However, this is only a very rough estimation and since $R_s\chi$ is positive, it could lead to a significant reduction in the magnitude of the Hall coefficient and probably offer an explanation for the observed decrease in the Hall coefficient on addition of 1 at. % Au.

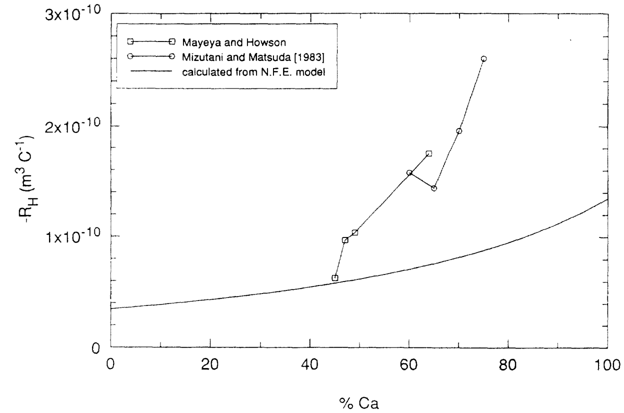

FIG. 3. Hall coefficient of $Ca_xAl_{100-x}$, including the data of Ref. 26 for comparison.

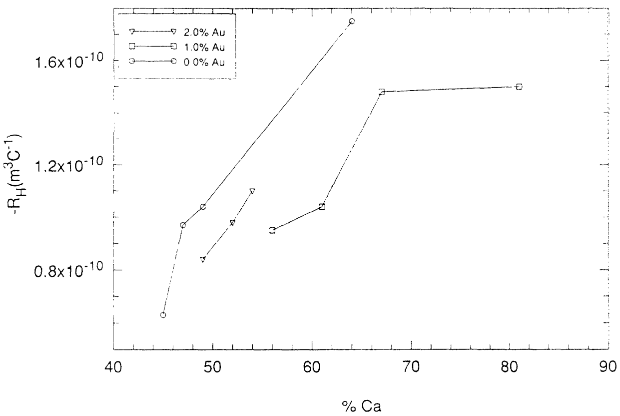

FIG. 4. Hall coefficient of $(Ca_xAl_{100-x})_{100-y}Au_y$, illustrating the effect of gold doping.

The reduction in $|R_H|$ is larger for 1 at. % Au than for 2 at. % Au. It is difficult to account for this observation quantitatively. However, it is consistent with the competing effects of the Au doping in the *s*-*d* hybridization model which increases $|R_H|$ as discussed above and the side-jump effect which reduces $|R_H|$.

The values of the Hall coefficient of CaAl(Au) were found to be independent of temperature as expected in systems in which the disorder scattering is isotropic. It has however, been argued that there exists a relationship

TABLE II. Side jump parameters.

| Alloy | $\rho$ ($\mu\Omega$ cm) | $\lambda_{so}$ (Å)$^2$ | $\tau_{so}$ (*ps*) | $R_s$ ($\mu m^3$ $C^{-1}$) |
|---|---|---|---|---|
| YAl | 260 | 0.71 | 0.7 | 1.0 |
| CaAl(Au) | 140–400 | 0.50 | 1.0 | 0.2–1.6 |

between the temperature dependence of the Hall coefficient $R_H$ and the variation in the electrical resistivity $\rho$, due to electron-electron interactions given as[29]

$$\frac{1}{R_H}\frac{dR_H}{dT}=\frac{2}{\rho}\frac{d\rho}{dT}\ . \tag{5}$$

In CaAl amorphous alloys, $\rho^{-1}d\rho/dT \sim 10^{-4}$ K$^{-1}$, which is too small to lead to a noticeable change in the Hall coefficient as a function of temperature. Thus, the effects of electron-electron interactions on the Hall coefficient in CaAl amorphous alloys are negligible. It has also been argued that the side-jump mechanism produces a temperature dependence of the Hall coefficient determined by the product $\rho^2\chi$.[9] In CaAl, $\chi$ has a temperature-independent value of $\sim 2.5\times10^{-4}$ JT$^{-2}$ (Ref. 26) and the change in the electrical resistivity is very small so that in the temperature range 1.5–2.5 K, $\rho^2\chi$ is almost constant. As a result the temperature dependence of the Hall coefficient in CaAl from the side-jump effect is also negligible.

## V. CONCLUSION

The Hall coefficients of CaAl(Au) amorphous alloys have been found to be negative and independent of temperature, which is easily understood as a direct consequence of isotropic scattering. However, their magnitudes are significantly higher than those for nearly-free-electron behavior. These deviations have been accounted for from considerations of the unusual electronic structure of CaAl, and by invoking the theory of *s*-*d* hybridization. *s*-*d* hybridization results from the presence of a significant number of *d* states at the Fermi level originating from the edge of the Ca *d* band. This interaction alters the dynamics of the *s* band and consequently affects the form of the Hall coefficient which depends on the density of states and the derivative of the density of states at the Fermi level.

The observed reduction in the magnitude of the Hall coefficient by gold doping has been argued to result from the side-jump effect. Although this effect has been used to account for the positive sign of the Hall coefficient in transition-metal alloys, its effect in CaAl(Au) is not sufficient to lead to a change in sign of the Hall coefficient, despite the large electrical resistivities of these alloys and strong spin-orbit scattering introduced by gold doping. Thus, it is apparent that a complete description of the Hall effect in CaAl(Au) must incorporate both the effects of *s*-*d* hybridization and the side-jump mechanism. *s*-*d* hybridization increases $|R_H|$ over the free-electron value and gold doping decreases the magnitude through the side-jump mechanism.

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