# Influence of spin-orbit scattering on the magnetoresistance due to enhanced electron-electron interactions

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We present a systematic study of the spin-orbit scattering effects on the low-temperature resistivity and magnetoresistance in a series of amorphous Ca-Al alloys. The level of the spin-orbit scattering was varied over a wide range from very weak to very strong by progressively adding either Ag or Au for Al. The data are analyzed within weak localization and enhanced electron-electron interaction theories. As the spin-orbit scattering increases, we find that the contribution to the magnetoresistance from electron-electron interaction diminishes to finally vanish, in the limit of extremely strong spin-orbit scattering. The spin-orbit scattering rate is found to increase linearly with the concentration up to 3 at. % of Au. The dephasing rate was also extracted and found to be consistent with electron-phonon scattering.

## I. INTRODUCTION

It is now well established that weak localization (WL) (Ref. 1) and enhanced electron-electron interactions (EEE) (Ref. 2) theories provide an accurate description of the low-temperature resistivity and magnetoresistance in a wide variety of disordered conductors. The two phenomena arise from quantum interference effects on the electron wave function when elastic scattering is extremely intense compared to all other scattering mechanisms. Their dependence on magnetic field, spin scattering, and inelastic scattering offer many interesting and complex features of the resistivity at low temperatures. Typically WL is readily observed and usually dominates the signal in these systems, whereas EEI effects require large magnetic fields and low levels of spin-spin and spin-orbit scattering in order to be detected. Moreover, their already small contribution to the magnetoresistance is strongly temperature dependent and decays rapidly with increasing temperature. This has led to difficulties in a quantitative determination of the EEI contribution to the magnetoresistance and has prevented conclusive tests of the theoretical predictions currently available. The EEI term is a combination of two terms: one is known as the diffusion channel term (DC) and the other as the Cooper channel term (CC). The former is characterized by an interaction between electrons with small *relative* momentum, whereas the latter involves pairs of electrons with small *total* momentum. The DC and CC terms are very sensitive to spin scattering (both spin-spin scattering and spin-orbit scattering). In a magnetic-impurity-free system, the CC contribution is expected to remain the same[2] while the DC contribution is expected to completely vanish in the limit of very strong spin-orbit scattering.[2-4] In the presence of high spin-spin scattering, on the other hand, both terms vanish in the same way as the WL contribution.

A recent study of the low-temperatures magnetoresistance in a series of free-electron-like amorphous alloys (Ca-Mg, Ca-Mg-Al, Ca-Al) by the authors[5,6] has shown that both WL and EEI terms are essential for describing the data. In particular at the lowest temperatures ($T \leq 4.2$ K) the WL contribution saturates so that the EEI part becomes clearly visible. Furthermore Ca-Mg-Al is the first three-dimensional system in which quantitative agreement between the theory and experiment was unambiguously found. Apart from being well characterized, the Ca-Mg-Al system has a very low level of spin-orbit scattering when compared with other amorphous systems. Hence the spin-orbit effect on EEI is at its minimum. This feature make the Ca-Mg-Al amorphous alloys very attractive if not unique for a quantitative study of the spin-orbit scattering effect on the magnetoresistance originating from EEI in bulk disordered conductors.

In this paper we present a study of the low-temperature magnetoresistance in a series of amorphous Ca-Al alloys where spin-orbit scattering has been systematically varied from very weak to very strong. This was achieved by progressively replacing Al with heavy elements (Ag and Au) which have a high spin-orbit coupling. The level of the dopant was kept low so that other electronic properties of the alloy are not appreciably affected. By this means, in an otherwise constant system the effect of spin-orbit scattering on the EEI magnetoresistance can be directly examined. Moreover this will also allow a test to the accuracy of the WL and EEI theories over a wide range of spin-orbit scattering.

## II. EXPERIMENTAL TECHNIQUES

The alloys were made by induction melting the appropriate amounts of the constituents under a high-purity argon atmosphere. The purity of the starting materials was Ca, 99.999%; Al, 99.9999%; Ag, 99.999%; Au, 99.9999%. Based on our earlier work, the concentration of magnetic impurities (in particular Mn, which is the only element expected to carry a moment in these alloys)

 

is estimated to be about 4 ppm or less.[5] The ribbons were prepared by melt spinning buttons of ~0.5 g onto a rotating Cu wheel with a tangential velocity of 42 m/s under 30 kPa of high-purity helium. The amorphous structure of the melt-spun ribbons was confirmed by x-ray diffraction using Cu $K\alpha$ radiation. The samples were then stored under liquid nitrogen to prevent crystallization and oxidation.

Table I summarizes the characteristics of the alloys. The mass density was measured by the Archimedes method with toluene as the working fluid and was found to increase linearly with Ag or Au concentration. The resistivity varies slightly with the alloy composition except at large concentrations of Ag and Au, where it decreases by up to approximately 30%. For $Ca_{70}Al_{30}$, the value of $\rho$ is in good agreement with that measured by Mizutani *et al.*[7] and with the value reported in our earlier work. The diffusivity $D$ for this alloy is calculated using the relation $D=1/e^2N(E_F)\rho$, where $N(E_F)$ is the density of states at the Fermi level as deduced from specific-heat measurements and $\rho$ is the measured resistivity. For the remaining alloys, $N(E_F)$ is assumed to stay constant since we do not expect it to change significantly upon substitution of Al by small amounts of Ag or Au. This assumption is supported by the results of Ca-Mg-Al,[7] where the density of states only changes by ~7% when going from $Ca_{70}Al_{30}$ to $Ca_{70}Mg_{10}Al_{20}$. At the same time the resistivity spans the same range of values as that observed in the present alloys. Thus the changes of the diffusivity in Table I are assumed to be a result of the changing resistivity only.

The magnetoresistance was measured using a four-probe ac bridge (LR400 from Linear Research, USA) in a standard liquid-helium cryostat. The power dissipated in the samples by the sensing current was kept low ($\leq 1$ $\mu$W) so that self-heating does not occur. Moreover, below 4.2 K the sample was directly immersed in the liquid. The temperature was monitored using a carbon-glass resistor and kept constant to within 1% during the magnetic-field sweeps. The magnetic field of up to 8.7 T was provided by a superconducting solenoid.

## III. RESULTS AND DISCUSSION

Representative magnetoresistance data of Ca-Al-$X$ alloys ($X$=Ag,Au) are shown in Fig. 1 at different temperatures between 1.5 and 30 K and in fields up to 8.7 T. The points are the experimental data and the solid line a fit as outlined below. In all samples, the magnetoresistance is positive at low fields reflecting the presence of spin-orbit scattering. At high fields, however, it is either positive or negative depending on the concentration of Ag or Au. For Au=2 and 3 at. %, the magnetoresistance is positive over the whole range of field and temperature, reflecting to the high degree of spin-orbit scattering in these alloys. As the temperature increases the overall magnitude of the magnetoresistance is reduced, due to increasing inelastic scattering which destroys the phase coherence of the electron wave function. Where they overlap, the data of $Ca_{70}Al_{30}$ are identical with our previous results.[5,6] In measurements reported by Tsai and Lu[8] on Ca-Al alloys and by Howson *et al.* on Ca-Al(Au),[9] the positive part of the magnetoresistance in the pure alloy (i.e., $Ca_{70}Al_{30}$) was not observed. This is characteristic of a relatively high amount of magnetic impurities, which also destroys phase coherence. This is of particular importance since it can lead to erroneous values of the spin-orbit scattering rate and the dephasing rate when quantitative analysis is made. In our alloys, the magnetic impurities level is low and its contribution to the magnetoresistance is negligible.[10]

The fitting procedure adopted here is the same as that followed in Ref. 5, the details being given by Baxter *et al.*[10] We start the fitting by restricting it to low fields, where only WL contributions to the magnetoresistance, keeping the spin-orbit field $B_{so}$ and the dephasing field $B_\phi$ as free parameters (the field $B_x$ is related to the scattering time $\tau_x$ through the relation $B_x=\hbar/4eD\tau_x$). In fact the fit is made more restrictive than this since $B_{so}$ must be independent of temperature. If we then extend the low-field fit to high fields, the theoretical curve from the WL expression lies below the experimental data. This difference is a direct reflection of the missing positive contribution from EEI. We therefore, as a second step, extend the fit to the entire field range including the EEI contribution with the only free parameter being the screening parameter $\tilde{F}_\sigma$. Only the DC contribution is retained since the CC term is very small in low diffusivity alloys, being about 5% of the total signal even at the lowest temperature (1.5 K and 8.7 T). The parameter $\tilde{F}_\sigma$ is related to the Coulomb interaction between the elec-

TABLE I. Physical parameters of Ca-Al-$X$ ($X$=Ag,Au) amorphous alloys.

| Alloy | $d$ (g/cm$^3$) | $\rho$ ($\mu\Omega$ cm) | $D$ (cm$^2$ s$^{-1}$) | $1/\tau_{so}$ ($10^{11}$ s$^{-1}$) | $\tilde{F}_\sigma$[a] | $\tilde{F}_\sigma$[b] |
|---|---|---|---|---|---|---|
| $Ca_{70}Al_{30}$ | 1.85±0.04 | 310±16 | 1.5 | 0.82±0.1 | 0.556±0.05 | 0.18±0.02 |
| $Ca_{70}Al_{29.7}Ag_{0.3}$ | 1.84±0.04 | 264±13 | 1.8 | 1.64±0.1 | 0.47±0.04 | 0.22±0.03 |
| $Ca_{70}Al_{29.3}Ag_{0.7}$ | 1.88±0.04 | 270±13 | 1.7 | 2.31±0.15 | 0.397±0.04 | 0.19±0.03 |
| $Ca_{70}Al_{28}Ag_2$ | 1.92±0.04 | 245±12 | 1.94 | 5.48±0.5 | 0.244±0.03 | 0.08±0.02 |
| $Ca_{70}Al_{29.9}Au_{0.1}$ | 1.86±0.04 | 297±15 | 1.56 | 6.28±1.0 | 0.185±0.02 | 0.015±0.02 |
| $Ca_{70}Al_{29.8}Au_{02}$ | 1.86±0.04 | 280±14 | 1.66 | 13.1±2 | 0.10±0.02 | 0.01±0.02 |
| $Ca_{70}Al_{29.6}Au_{0.4}$ | 1.87±0.04 | 280±14 | 1.66 | 24.2±4.0 | 0.075±0.02 | −0.084±0.02 |
| $Ca_{70}Al_{29.2}Au_{0.8}$ | 1.92±0.04 | 290±15 | 1.6 | 48.6±10 | 0.015±0.02 | −0.081±0.02 |
| $Ca_{70}Al_{28}Au_2$ | 2.00±0.04 | 220±14 | 2.1 | 70.5±15.0 | 0.0±0.02 | −0.106±0.02 |
| $Ca_{70}Al_{27}Au_3$ | 2.12±0.04 | 210±14 | 2.27 | 152±35.0 | 0.0±0.02 | −0.246±0.02 |

[a]From magnetoresistance.

[b]From the temperature dependence of the resistivity.

trons and depends on the details of the electron screening and the Fermi surface of the conductor under consideration. For free-electron systems in three dimensions, $\tilde{F}_\sigma$ has been given as[2,3]

$$\tilde{F}_\sigma = -\frac{32}{3F}[1+\tfrac{3}{4}F-(1-F/2)^{3/2}] , \quad (1)$$

where $F$ is the average over the Fermi surface of the screened Coulomb interaction. For $F \ll 1$, $\tilde{F}_\sigma \approx F$.

Like $B_{so}$, $\tilde{F}_\sigma$ is also temperature independent. Hence the final theoretical curves in Fig. 1 and for the remaining alloys are generated with common values of $B_{so}$ and $\tilde{F}_\sigma$ and a value of $B_\phi$ at each temperature for each family of curves. The agreement between the theory and experiment is excellent over the full range of field and temperature. Furthermore, the quality of the fit is equally good for all alloys irrespective of their composition as shown in Fig. 2. Thus our first significant conclusion is that the quantum corrections to the conductivity expressions (QCC) give a very good description of the data in this alloy system regardless of the level of the spin-orbit scattering. We therefore conclude that WL and EEI theories give the right field and temperature dependence of the magnetoresistance not only over a wide range of $B/T$ as found earlier,[6] but also over a wide range of spin-orbit scattering.

The behavior of the dephasing rate $1/\tau_\phi = 4eDB_\phi/\hbar$ as a function of temperature is shown in Fig. 3. All samples give essentially the same value, hence providing a further consistency check to our results. The temperature depen-

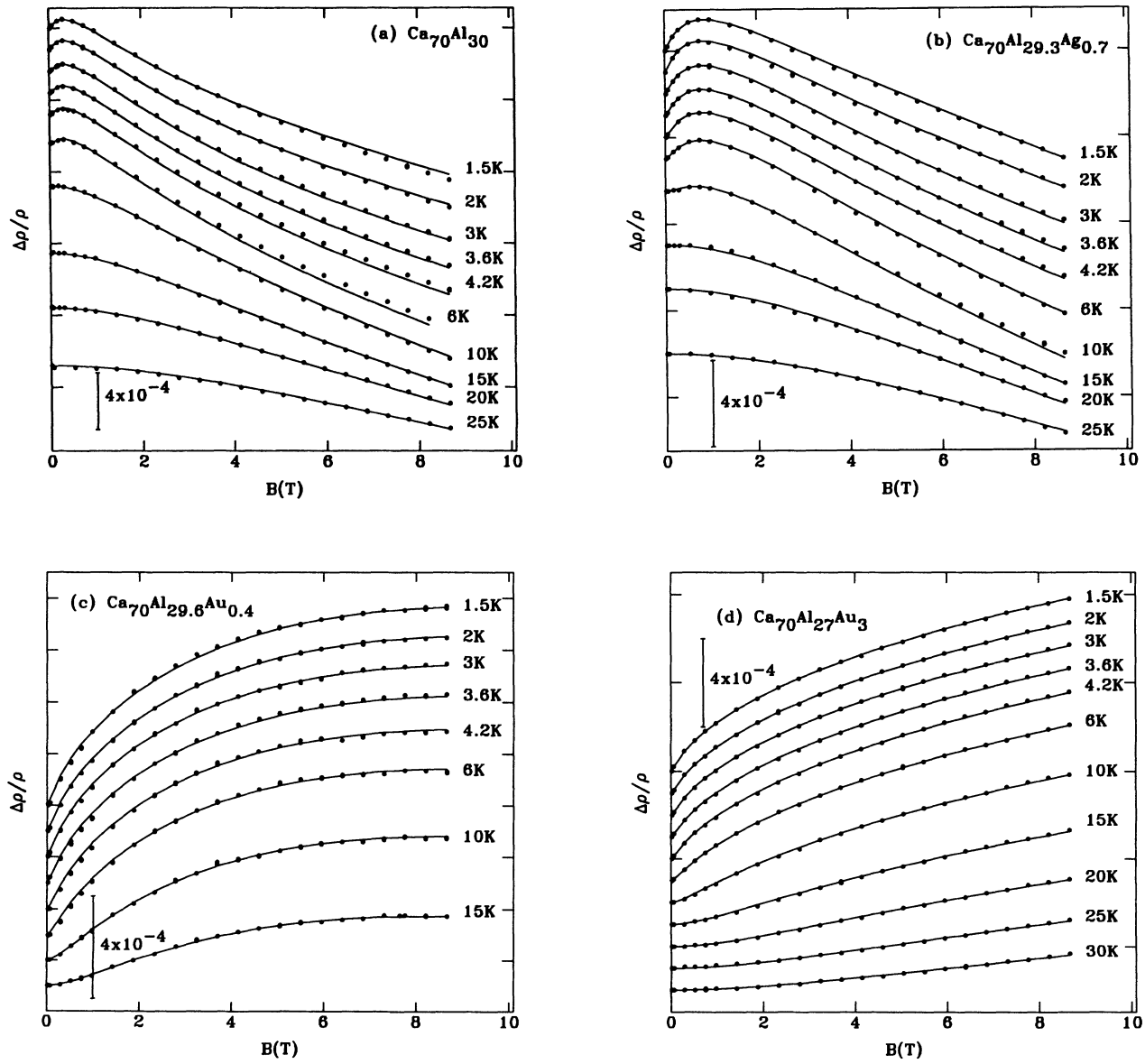


FIG. 1. Magnetoresistance of (a) $Ca_{70}Al_{30}$, (b) $Ca_{70}Al_{29.3}Ag_{0.7}$, (c) $Ca_{70}Al_{29.6}Au_{0.4}$, and (d) $Ca_{70}Al_{27}Au_3$ at different temperatures. The points are the experimental data and the solid line is a fit as described in the text.

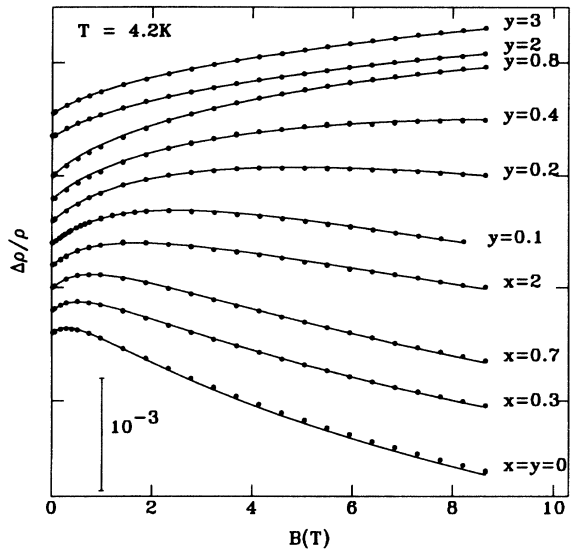


FIG. 2. Magnetoresistance of amorphous Ca-Al-$X$ ($X$=Ag,Au) alloys at 4.2 K. The points are the experimental data and the solid line a fit as described in the text. The Ag ($x$) and Au ($y$) concentrations are indicated in the figure.

dence may be described by an expression of the form $1/\tau_\phi = 1/\tau_\phi^0 + AT^p$ with $p = 3.0 \pm 0.3$. Similar behavior has also been reported by other workers using different materials but in some cases their results give smaller values of the exponent $p$. In Mg films[11] and in various metallic glasses[12,13] $1/\tau_\phi$ varies as $T^2$. According to Chakravarty and Schmid,[14] the exponent $p$ can take any value between 2 and 4 depending on the different phonon modes involved in the scattering process. Thus we can conclude that dephasing in amorphous Ca-Al-$X$ ($X$=Ag,Au) alloys is due to inelastic electron-phonon scattering in three dimensions. The saturation value $1/\tau_\phi^0$ may be due to residual spin scattering from magnetic impurities present in the alloys, although a similar saturation of $\tau_\phi$ has also been observed in systems where the magnetic impurity level was too small to account for this behavior.[15]

The spin-orbit scattering rate $1/\tau_{so} = 4eDB_{so}/\hbar$ as deduced from the fitting is shown in Fig. 4 against Ag and Au concentrations. The large error bars at 2 and 3 at. % of Au are due to the fact that the relatively featureless magnetoresistance data at these concentrations are relatively insensitive to the exact value of $1/\tau_{so}$. Where spin-orbit scattering is low the sign change in the magnetoresistance with magnetic field defines $\tau_{so}$ much more precisely. The spin-orbit scattering rate changes by more than two orders of magnitude from $8.2\times10^{10}$ to $1.52\times10^{13}$ s$^{-1}$. The value for $Ca_{70}Al_{30}$ is consistent with our previous results. However, the value reported by Howson *et al.*[9] is significantly different ($\sim10^4$ s$^{-1}$). This unphysically low value is probably an artifact of a high level of magnetic impurities which suppress the positive part of the magnetoresistance at low field as mentioned above. In fact, using our value of $1/\tau_{so}$ we find that in order to fit their data $1/\tau_\phi$ must take a value of $\approx 1.7\times10^{11}$ s$^{-1}$. From this we can infer a magnetic impurity contamination (for example by Mn) of about 50 ppm in the samples used in Ref. 9, which underscores the need to use materials of the highest purity for studying the QCC in disordered conductors. It is also interesting to note that even at the largest concentration of Au, the condition $1/\tau_e \gg 1/\tau_{so}$ (where $\tau_e$ is the elastic scattering time) under which the WL expression was derived is still satisfied[16] since $1/\tau_e \sim 10^{15}$ s$^{-1}$. It may be seen from the figure that the spin-orbit scattering rate increases linearly

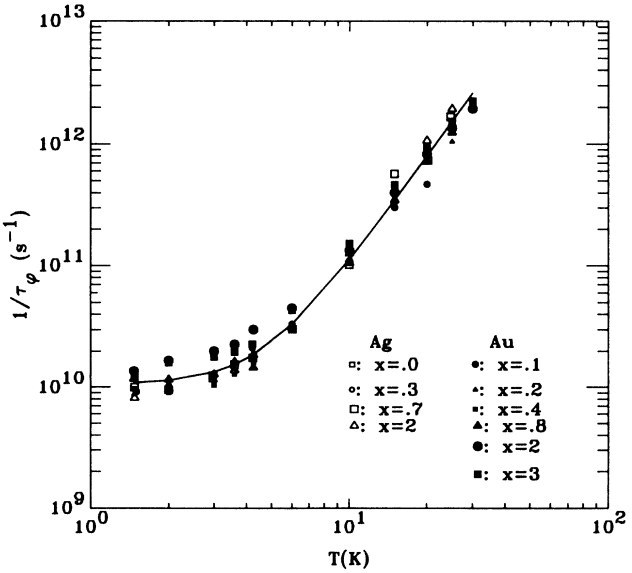


FIG. 3. Dephasing rate $1/\tau_\phi$ as a function of temperature in amorphous Ca-Al-$X$ ($X$=Ag,Au) alloys. The solid line is a fit using expression $1/\tau_\phi = 1/\tau_\phi^0 + AT^p$ with $p=3$.

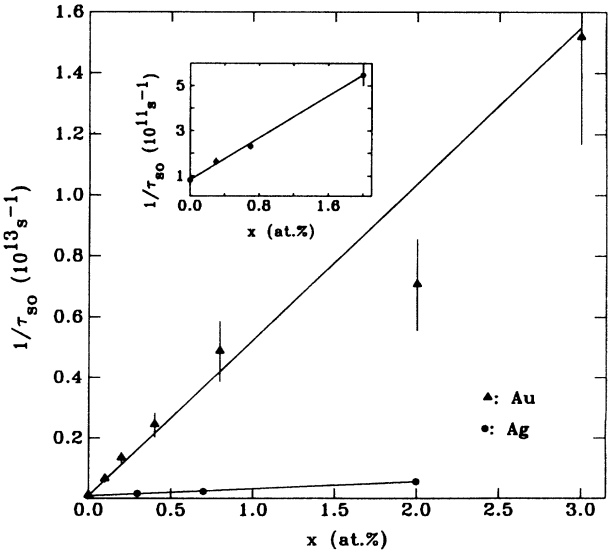


FIG. 4. The spin-orbit scattering rate as a function of the Ag and Au concentration in amorphous Ca-Al-$X$ ($X$=Ag,Au) alloys. Inset: $1/\tau_{so}$ for $Ca_{70}Al_{30-x}Ag_x$ on an enlarged scale. The solid line is a guide to the eye.

with the concentration; however, the increase is much faster for Au content than for Ag, as expected. The ratio between the two slopes is $\approx 21$. A similar result was found by Richter, Baxter, and Ström-Olsen[15] in Mg-Cu and Mg-Zn amorphous alloys also doped with Ag and Au over the same concentration range, although the absolute value of the spin-orbit scattering rate at a given Au or Ag content is larger in the present case. If the atomic orbital wave-functions approach is assumed, the spin-orbit scattering rate should scale as $Z^8$ where $Z$ is the atomic number.[17] Applying this to the present case we get $(Z_{Au}/Z_{Ag})^8 \approx 63$, which is significantly different from what we obtained above. Instead our value and that of Ref. 15 are very close to $(Z_{Au}/Z_{Ag})^6 \approx 22.5$. We have no explanation for this weaker dependence except that perhaps it reflects the screening of the ionic charge by the electrons.

The most significant point that emerges from our analysis is the suppression of the DC contribution to the magnetoresistance by spin-orbit scattering, as predicted by the theory. To our knowledge this is the first time where such an observation has been quantified. The magnetic-field-dependent part of the DC correction to the resistivity is due to the interaction between electrons with a total spin moment $j=1$ and $M=\pm 1$.[4,18] In the presence of spin scattering (spin-orbit and/or spin-spin scattering), this magnetic-field dependence is suppressed when the Zeeman splitting $g\mu_B B \ll \hbar/t_s$.[4] $t_s$ is the total spin relaxation time and is given by $1/t_s = \frac{4}{3}(1/\tau_{so} + 1/\tau_s)$, where $1/\tau_s$ is the spin-spin scattering rate. Since $1/\tau_s$ is assumed to be constant ($\approx 1/\tau_\phi^0 = 10^{10}\ s^{-1}$) the equality in the above condition is reached for $1/\tau_{so} \approx 1.1\times 10^{12}\ s^{-1}$. Figure 5 shows that at this scattering rate the DC contribution is already reduced to about a fifth of its low spin-orbit scattering value. Further increase in $1/\tau_{so}$ results in a complete removal of the effect. We have used the screening parameter $\tilde{F}_\sigma$, since it appears as a scaling factor in the DC magnetoresistance expression, as a measure of the progressive reduction in the DC contribution. It is worth mentioning that in $Ca_{70}Al_{30}$ the value of $\tilde{F}_\sigma$ is very close to the free-electron value $\tilde{F}_\sigma^0 \approx 0.52$ estimated using the Thomas-Fermi theory, and is therefore another indication that in this alloy we are in the limit of very weak spin-orbit scattering.

Figure 6 shows the temperature dependence of the resistivity between 1.5 and 4.3 K for six different compositions. For clarity the data of the other alloys are not included in the graph. The data are plotted as $\Delta\rho\sqrt{D}/\rho^2$ against $\sqrt{T}$ so that the effect of spin-orbit scattering can be clearly displayed. The solid line is a fit using the DC expression of the resistivity temperature dependence given by[3]

$$\frac{\Delta\rho}{\rho^2} = -\frac{0.915e^2}{4\pi^2\hbar}\left(\frac{4}{3}-\frac{3}{2}\tilde{F}_\sigma\right)\left[\frac{k_B T}{\hbar D}\right]^{1/2}. \tag{2}$$

The WL localization contribution is very small and can be neglected since the temperature dependence to the resistivity comes through the dephasing time $\tau_\phi$, which is constant. As for the magnetoresistance, the fit is excellent for all alloys and over the full temperature range. However, the best fits are obtained for smaller values of the screening parameter $\tilde{F}_\sigma$ than those found from the magnetoresistance analysis. Moreover, for large spin-orbit scattering alloys $\tilde{F}_\sigma$ has to be negative in order to fit the data (see Table I). This is not an isolated case and in fact Poon, Wong, and Drehman[19] have found that it was necessary for $\tilde{F}_\sigma$ to take a negative value ($-0.6$) in order

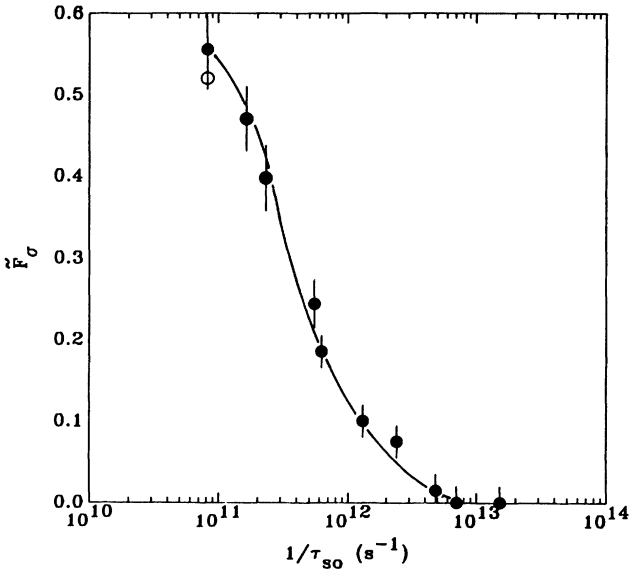


FIG. 5. The diffusion channel contribution to the magnetoresistance ($\propto \tilde{F}_\sigma$) as a function of the spin-orbit scattering rate. The open circle is the estimated free-electron value for $Ca_{70}Al_{30}$. The solid line is a guide to the eye.

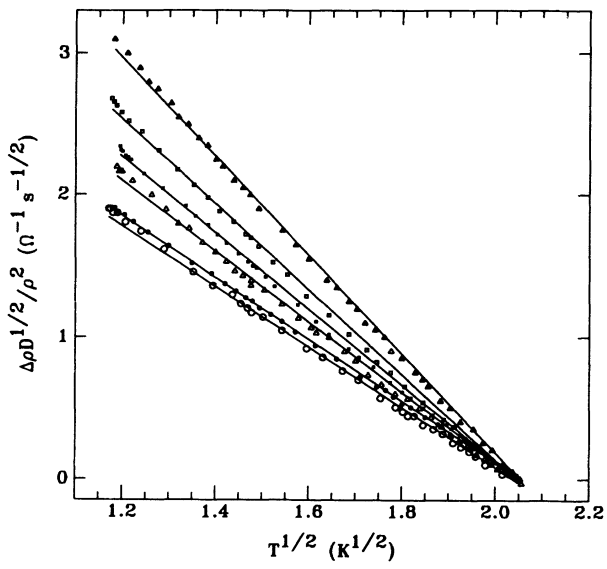


FIG. 6. Low-temperature resistivity of amorphous Ca-Al-$X$ ($X$=Ag,Au) alloys. The points are the experimental data and the solid line a fit using Eq. (2). (●) $Ca_{70}Al_{30}$, (○) $Ca_{70}Al_{29.3}Ag_{0.7}$, (△) $Ca_{70}Al_{28}Ag_2$, (■) $Ca_{70}Al_{29.9}Au_{0.1}$, (□) $Ca_{70}Al_{29.2}Au_{0.8}$, (▲) $Ca_{70}Al_{27}Au_3$.

to account for the temperature dependence of the resistivity of amorphous Lu-Pd and Lu-Ni alloys, which are characterized by a strong spin-orbit scattering ($\sim 10^{14}$ $s^{-1}$) about a factor 10 larger than that of $Ca_{70}Al_{27}Au_3$. Furthermore it has also been found that $\tilde{F}_\sigma \approx -0.05$ in amorphous Cu-Zr (Ref. 19) where the spin-orbit scattering rate is $\approx 1.2\times 10^{12}$ $s^{-1}$ (one should note that in order to be consistent with our definition of $\tau_{so}$ the values given in Ref. 19 should be multiplied by 3). This value is very close to that at which we observe the change of sign in our samples and is therefore a further support to our results. According to Al'tshuler, Aronov, and Zuzin,[4] the $\frac{3}{2}\tilde{F}_\sigma$ term in Eq. (2) should be replaced by $\frac{3}{8}\tilde{F}_\sigma$ in the limit of high spin-orbit scattering. However, even with this adjustment it is not possible to account for the observed magnitude of the resistivity at large spin-orbit scattering rates. On the other hand, it was also suggested, when attractive interaction between electrons via virtual phonons is included, that the Coulomb interaction constant $F$ should be replaced by $F^*=F-2\lambda$, $\lambda$ being the electron-phonon mass-enhancement parameter.[2,17,20] In this case negative values of $\tilde{F}_\sigma$ are allowed when $F$ is small ($<2\lambda$). But though this redefinition of $F$ might explain the reduced values of $\tilde{F}_\sigma$, it certainly fails to account for the systematic change with the spin-orbit scattering rate. Therefore we are led to the conclusion that the above equation needs to be refined and in particular should explicitly include the effect of the spin-orbit scattering.

## IV. CONCLUSION

We have shown that WL and EEI theories give an accurate description of the magnetic field and temperature dependence of the resistivity of amorphous Ca-Al-$X$($X$=Ag,Au) alloys over a wide range of spin-orbit scattering. From the discussion we have also shown how the presence of magnetic impurities significantly alters the magnetoresistance and therefore can lead to erroneous results. Furthermore, we have presented evidence of the effect of spin-orbit scattering on the EEI term originating from the DC. We found that its contribution to the magnetoresistance decreases rapidly with increasing spin-orbit scattering and is totally suppressed in the extremely high spin-orbit scattering regime in agreement with the theoretical predictions. Finally, we have also shown that in its actual form, the temperature-dependence expression of the resistivity from the DC cannot account for the observed magnitude of the resistivity change in high spin-orbit scattering alloys unless negative values of the screening parameter are assumed.

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