Weak-localization and Coulombic interaction effects in the low-temperature resistivity and magnetoresistivity of Y-Al metallic glasses

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We report measurements of the electrical resistivity and magnetoresistivity of the nonsuperconducting and nonmagnetic metallic glass $Y_{1-x}Al_x$ ($0.2 \leq x \leq 0.4$) from 60 mK to 17 K and in magnetic fields up to 4.3 T. We find that weak localization and Coulombic interaction provide a good quantitative description of the temperature dependence of the electrical resistivity but only a semiquantitative description of the magnetoresistivity.

Interest in the low-temperature electrical resistivity of metallic glasses has been rekindled by two theories of the quantum corrections to the conductivity: weak localization and Coulombic interaction. Between them, these theories[1] appear to offer a natural explanation of two striking features of metallic glasses: the temperature coefficient of resistivity, which (in the absence of superconductivity) is always negative at sufficiently low temperatures, and the magnetoresistivity, which is far larger than any "normal" contributions due to the Lorentz force. The temperature dependence gives a $-\sqrt{T}$ behavior below about 3 K,[2] as predicted by the Coulombic interaction model. The interaction model has also been used to interpret magnetoresistance in Cu-Ti (Ref. 3) and La-Al (Ref. 4) glasses, but the interpretation does not stand up to quantitative analysis (as has been recognized by the authors of Ref. 3). Instead, weak localization seems to give a better description of the magnetoresistance and, in fact, several fittings based on weak localization (with additional corrections where appropriate for superconducting fluctuations) have been presented for magnetoresistance in Cu-Zr,[5] Ca-Al,[6] Y-Al,[7] and Lu-Pd.[8] The magnitude of the effect is well described, but there are discrepancies in the detailed field or temperature dependence.

In the present article we report measurements of resistivity and magnetoresistivity on $Y_{1-x}Al_x$ ($0.2 \leq x \leq 0.4$) metallic glasses down to 60 mK and in fields up to 4.3 T. The results are tested in detail against the predictions of both weak localization and Coulombic interactions. Y-Al was chosen because it shows no detectable superconductivity or magnetic order leading to less ambiguity in identifying contributions to the transport properties.

The alloys were prepared by melt spinning 1-g buttons of Y-Al onto a copper wheel having a tangential velocity of about 50 m/s. The buttons were prepared by arc melting appropriate amounts of Y of 99.95% purity and Al of 99.999% purity. The resulting ribbons were found to be amorphous through x-ray diffraction.[9] The electrical resistance was measured by a four-terminal ac technique to within an accuracy of a few parts in 10^6. In the temperature range 1.3–4.2 K, the magnetoresistance was measured in longitudinal field up to 4.3 T, above 4.2 K in a transverse field up to 1 T.

The resistivity ρ and its temperature coefficient $\alpha = \partial \ln\rho / \partial T$ of Y-Al glasses show little variation with composition (Table I). In fact, the temperature dependence of ρ is essentially identical in all our samples. A representative plot of the behavior below about 10 K is given in Fig. 1.

TABLE I. Electrical resistivity ρ(290 K), mass density ρ_m, resistivity ratio $\rho(290)/\rho(0)$, and room-temperature resistivity coefficient α_{290} of YAl metallic glasses.

System	ρ_m(g cm^{-3})	ρ(290 K)($\mu\Omega$cm)	$\rho(290)/\rho(0)$	α_{290}(10^{-4} K^{-1})
$Y_{54.0}Al_{46.0}$	3.97 ±0.06	253 ±9	· · ·	−0.80 ±0.2
$Y_{60.0}Al_{40.0}$	3.99 ±0.06	257 ±9	0.95(0)	−0.95 ±0.2
$Y_{62.5}Al_{37.5}$	4.02 ±0.06	266 ±9	· · ·	−0.97 ±0.2
$Y_{65.0}Al_{35.0}$	4.06 ±0.06	259 ±9	· · ·	−0.92 ±0.2
$Y_{66.7}Al_{33.3}$	4.17 ±0.06	268 ±9	0.94(9)	−0.96 ±0.2
$Y_{68.0}Al_{32.0}$	4.11 ±0.06	275 ±9	· · ·	−1.27 ±0.2
$Y_{70.5}Al_{29.5}$	4.16 ±0.06	260 ±9	· · ·	−1.03 ±0.2
$Y_{72.6}Al_{27.4}$	4.25 ±0.06	259 ±9	· · ·	−1.04 ±0.2
$Y_{75.0}Al_{25.0}$	4.25 ±0.06	259 ±9	· · ·	−1.25 ±0.2
$Y_{77.5}Al_{22.5}$	4.26 ±0.06	275 ±9	0.94(6)	−1.14 ±0.2
$Y_{80.0}Al_{20.0}$	4.27 ±0.06	262 ±9	· · ·	−1.37 ±0.2

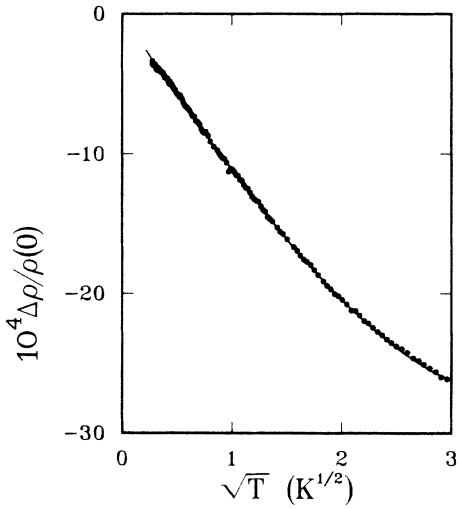

FIG. 1. Normalized resistivity of $Y_{77.5}Al_{22.5}$ plotted as a function of $\sqrt{T}$. The solid line is a least-squares fit to the data calculated as described in the text.

From about 2.5 K down to the lowest temperature obtainable in our dilution refrigerator (≈ 60 mK) ρ varies as $-\sqrt{T}$, as also reported for $Y_{75}Al_{25}$.[8] A $\sqrt{T}$ variation is predicted by the Coulombic interaction model.

The principal contribution to the resistivity due to Coulombic interactions arises from the so-called diffusion channel, which predicts (in the absence of any spin-orbit interaction) a correction to the conductivity σ_D,[1]

$$\sigma_D(H,T)=\frac{1.3}{\sqrt{2}}\left(\frac{e^2}{4\pi^2\hbar}\right)\left(\frac{k_BT}{\hbar D}\right)^{1/2}\times\left[\frac{4}{3}+\lambda^{J=1}(F)\left(\frac{3}{2}+\frac{g_3(h)}{1.3}\right)\right], \quad (1)$$

where $D=\sigma/[e^2(dN/d\mu)]$ is the diffusion constant of the conduction electrons, $dN/d\mu$ is the electronic density of states, $\lambda^{J=1}(F)$ is a parameter dependent on the strength of the electron-electron interaction, and $h=g\mu_BH/k_BT$. The function $g_3(h)$ goes asymptotically to $0.056h^2$ when $h\ll 1$ and to $(\sqrt{h}-1.3)$ when $h\gg 1$. In Y-Al we may expect there to be significant spin-orbit scattering. No theory for $\sigma_D(H,T)$ at present includes simultaneously both temperature and spin-orbit scattering, but when $\hbar\tau_{so}^{-1}\gg k_BT$ (τ_{so}^{-1} being the spin-orbit scattering rate) the contribution from $\lambda^{J=1}(F)$ becomes negligible. Below 3 K this criterion is well satisfied in Y-Al, as we shall see later. Putting $\lambda=0$ in (1) gives $D=5\times10^{-5}$ m^2/s. From the overall magnitude of the conductivity we find $D\geq 10^{-4}$ m^2/s, using photoemission data[10] for the electronic density of states at E_F. The factor of about 2 between the two values may not be significant. There is some uncertainty in the value of $dN/d\mu$; also (1) results from a perturbation expansion valid when $(k_Fl_e)^{-1}\ll 1$, l_e being the elastic scattering length and for Y-Al we find $(k_Fl_e)^{-1}\approx 0.3$, so that higher terms may need to be considered.[11,12]

Weak localization—a one-electron interference effect arising from intense elastic scattering—gives a term which may be written

$$\sigma_L(0,T)=\frac{e^2}{\pi^2\hbar}\left(\frac{e}{4\hbar}\right)^{1/2}\times\left[(H_\phi+2H_s)^{1/2}-3\left(H_\phi+\frac{2H_s}{3}+\frac{4H_{so}}{3}\right)^{1/2}\right], \quad (2)$$

where $H_l=\hbar/(4eD\tau_l)$ and τ_ϕ, τ_{so}, and τ_s are the phase-coherence, spin-orbit, and spin-scattering times, respectively. The temperature dependence comes mainly from H_ϕ, which increases with T as the inelastic scattering rate increases. When spin scattering is small ($H_s\ll H_\phi,H_{so}$) (2) predicts σ_L to increase with T when $H_\phi\leq 3H_{so}/8$ and to decrease with T when $H_\phi\geq 3H_{so}/8$. As we shall show from the analysis of our magnetoresistance data, we believe that $H_{so}\gg H_\phi$ for $T\leq 10$ K, so the contribution to the resistivity from weak localization is positive. Since one expects (for electron-phonon scattering) $\tau_\phi\propto T^2$, σ_L should vary as T. Finally, one expects a contribution to ρ from the temperature dependence of the structure factor. To lowest order[13] this should vary as T^2. We have therefore fitted the data to an expression $\Delta\rho/\rho=a\sqrt{T}+bT+cT^2$, the fit being the line through the points in Fig. 1. It is not surprising that a three-parameter fit through such slowly varying data should work so well. However, the value of the fitting parameters may be examined to see if they are physically reasonable. The parameter $a\simeq -1.62\times10^{-3}$ K$^{-1/2}$ is the slope of the $\sqrt{T}$ region, and is, as we have discussed, consistent with the interaction model. The parameter b takes the value 6.1×10^{-5} K^{-1}; using weak-localization parameters from a best fit to the magnetoresistance (see below) we estimate $b\simeq 9\times10^{-5}$ K^{-1}. The parameter c is 5.3×10^{-6} K^{-2}, which corresponds[13] to a Debye temperature of about

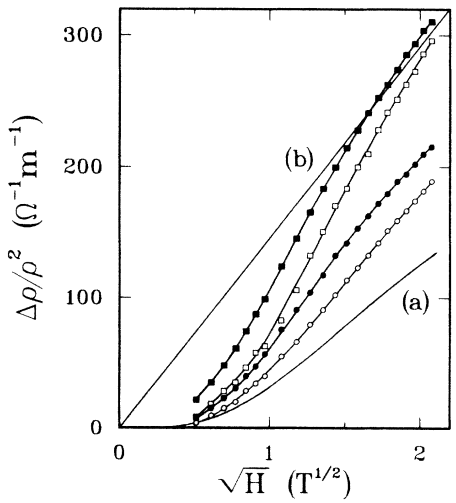

FIG. 2. Normalized magnetoresistivity $\Delta\rho/\rho^2$ of $Y_{60}Al_{40}$ and $Y_{80}Al_{20}$ (○ and □) plotted as a function of $\sqrt{H}$, at 1.2 K (solid symbols), and 4.2 K (open symbols). The solid lines through the data are guides to the eye. The curve marked (a) has been calculated from Eq. (4) using $H_{so}=3$ T, $D=10^{-4}$ m^2/s, and $H_\phi\simeq 0.187$ T T (estimated from the low-field magnetoresistance data of $Y_{60}Al_{40}$ at 4.2 K as described in the text). The curve marked (b) represents the upper limit for the weak-localization contribution ($H_\phi=0$ and $H_{so}\rightarrow\infty$).

150 K, a quite feasible value for these alloys. The fact that the parameters are reasonable not only lends credibility to the model, but also indicates that above about 3 K all three terms make a measurable contribution to ρ, but that below 3 K the Coulombic interaction term dominates.

The situation is less satisfactory when the magnetoresistivity is considered. Figure 2 shows $\rho(H,T)-\rho(0,T)/\rho^2$ plotted against $\sqrt{H}$ for two samples of Y-Al up to fields of 4.3 T at the two temperatures 1.2 and 4.2 K. The data show a characteristic variation[5-8] as H^2 at low field giving way to $\sqrt{H}$ at high field. In the range 1.2–4.2 K there is little temperature dependence. However, above 4.2 K a much stronger temperature dependence is seen, as is shown in Fig. 3. The magnitude of the magnetoresistivity is inconsistent with the prediction of Eq. (1). If we calculate the contribution neglecting spin-orbit coupling using $\lambda^{J=1}\simeq -0.5$ (estimated from the averaged statically screened Coulomb interaction[1]), the predicted magnetoresistivity is almost zero on the scale of Fig. 2 at 4.2 K and not more than a third of the observed signal at 1.2 K. When spin-orbit scattering is considered the contribution is reduced still further. If $H \ll H_{so}^{i}=\hbar\tau_{so}^{-1}/g\mu_B$, then the Zeeman-splitting magnetoresistivity remains in its low-field limit and is given by

$$\Delta\sigma_D(H)\simeq -\frac{0.01e^2F}{\pi^2\hbar}\left(\frac{g\mu_B H_{so}^{i}}{\hbar D}\right)^{1/2}\left(\frac{H}{H_{so}^{i}}\right)^2 . \tag{3}$$

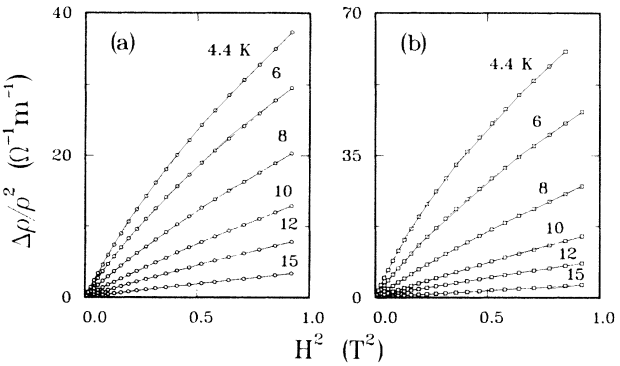

FIG. 3. Normalized magnetoresistivity $\Delta\rho/\rho^2$ plotted as a function of H^2 and at various temperatures: (a) $Y_{60}Al_{40}$, (b) $Y_{80}Al_{20}$.

In the present work this term is negligible.

A similar observation applies to the density-of-states correction,[1] which we have evaluated using the same value of the parameters as earlier. Even under the worst conditions (1.2 K and 4.3 T) the contribution is only about 7% of the observed signal.

However, weak localization makes a large contribution to the magnetoresistivity. The change in conductivity from this source, $\Delta\sigma_L=\sigma_L(H,T)-\sigma_L(0,T)$, including Zeeman splitting, spin-orbit interactions, and superconducting fluctuations, may be written as[14]

$$\Delta\sigma_L=\frac{e^2}{2\pi^2\hbar}\left(\frac{eH}{\hbar}\right)^{1/2}\left\{f_3\left(\frac{H_2}{H}\right)-c_{so}\beta f_3\left(\frac{H_\phi}{H}\right)+\frac{0.5}{\sqrt{1-\gamma}}\left[f_3\left(\frac{A_+}{H}\right)-f_3\left(\frac{A_-}{H}\right)\right]\right\}+B\left\{\frac{-1}{\sqrt{1-\gamma}}(\sqrt{t_-}-\sqrt{t_+})+\sqrt{t}-\sqrt{t+1}\right\} , \tag{4}$$

with

$$B=\frac{e^2}{\sqrt{3}\pi^2\hbar}\left(\frac{eH_{so}}{\hbar}\right)^{1/2} , \tag{4a}$$

$$\gamma=(3H/2H_{so}^{i})^2 , \tag{4b}$$

$$A_\pm=H_\phi+(2H_{so}/3)(1\pm\sqrt{1-\gamma}) , \tag{4c}$$

$$t_\pm=t+0.5(1\pm\sqrt{1-\gamma});\quad t=(3H_\phi/4H_{so}) . \tag{4d}$$

In Eq. (4) H_2 has been written for $H_\phi+4H_{so}/3$. The Maki-Thompson coefficient β has been included for completeness, but is negligible here ($\beta\sim 0.01$), because Y-Al is not superconducting. In the low-field limit ($H \ll H_{so}$, H_ϕ, $g\mu_B/k_BT$, H_{so}^{i}, and $\pi k_BT/2eD$) Eq. (4) takes the form $\Delta\rho/\rho^2\simeq\alpha H^2$.

From Fig. 3 we can extract the experimental value of α for different temperatures. Figure 4 shows a log-log plot of α against T. Above 4 K, α varies roughly as T^{-3}, but below 4 K it varies much more slowly and, in fact, appears to saturate.

The low-field form of Eq. (4) gives

$$\alpha\simeq\left(\frac{e^2}{4\pi^2\hbar}\right)\left(\frac{e}{\hbar H_{so}^3}\right)^{1/2}\times\left[\frac{1}{48}[y^{-3/2}-3(\tfrac{4}{3}+y)^{-3/2}]+\frac{c}{\sqrt{3}}\left(\frac{1}{4\sqrt{3y}}-\frac{3}{8}\right)\right] , \tag{5}$$

where $c=(3H_{so}/2H_{so}^{i})^2$ and $y=H_\phi/H_{so}$. Because α varies as T^{-3} above 4 K, it is believed that only one term from Eq. (5) makes a significant contribution in this region. In Y-Al we estimate $H_{so}^{i}\simeq 3.5H_{so}$, so that α should become negative when $H_\phi/H_{so}\geq 0.6$ and vary as $H_\phi^{-3/2}$ at low temperature. Assuming that we are in the low-temperature regime ($y \ll 1$), we estimate that above 4 K $\tau_\phi^{-1}\simeq 2.6\times10^{-9}T^2$ s^{-1}. Although τ_ϕ^{-1} is not exactly equal to

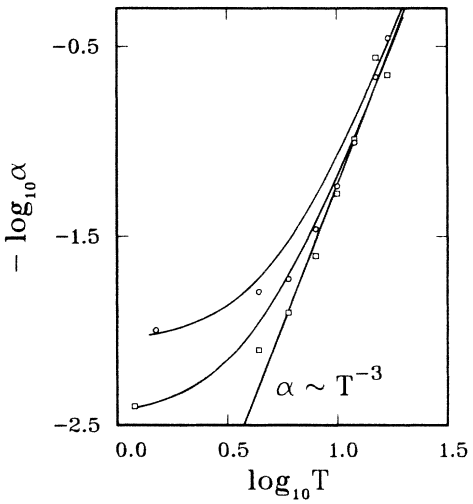

FIG. 4. Low-field magnetoresistivity coefficient $-\log_{10}\alpha$ plotted as a function of $\log_{10}T$: ○, $Y_{60}Al_{40}$; □, $Y_{80}Al_{20}$. The solid lines through the data are fits to $\alpha\sim 5(a+bT^2)^{-3/2}$ as described in the text.

τ_{in}^{-1}, the latter may be used to provide an estimate of the former. A simple estimate of the low-temperature electron-phonon scattering rate in disordered metals[15] gives $\tau_{in,e\text{-}ph}^{-1} \simeq 4\times 10^9 T^2$ s^{-1} close to the observed behavior of τ_ϕ^{-1}. The electron-electron phase-coherence rate in the present temperature range is smaller and has a leading term varying as T at low temperature.[16] It therefore appears that the behavior of τ_ϕ^{-1} above 4 K is controlled by electron-phonon scattering, as has been suggested for other glasses.[6,8,9] The absence of any significant deviation from $\alpha \sim T^{-3}$ even at 20 K allows us to give a lower bound for H_{so} of about 3 T (i.e., $\tau_{so} \leq 7\times 10^{-13}$ s), a value consistent with estimates made for its neighbor in the Periodic Table, Zr.[5,7,8] Below 4 K another mechanism must be responsible for the loss of phase coherence. There must be a crossover at some temperature to electron-electron controlled scattering which would give $\alpha \sim T^{-3/2}$. However, the actual behavior of α gives $H_\phi \sim (a + bT^2)$ rather than $cT + bT^2$, suggesting a temperature-independent mechanism. It is noteworthy that saturation of τ_ϕ^{-1} is seen quite widely in disordered metal systems.

One explanation—that the effect is caused by spin scattering from magnetic impurities in the yttrium (for example, a few hundred ppm of Gd would suffice)—is invalidated by the fact that a (0.13 T in $Y_{60}Al_{40}$ and 0.067 T in $Y_{80}Al_{20}$) scales with the Al concentration rather than the Y concentration.

Further difficulties arise when we try to describe the behavior at 4.2 and 1.2 K over the full-field range. To do this requires a value for H_{so}. If we use the value of 3 T, the calculated value of $\Delta\sigma_L$ is too small at high fields, as shown in Fig. 2. The value of the magnetoresistance can be increased by increasing H_{so}, but even if we allow H_{so} to be a free parameter we still cannot fit the measured data. This is dramatically illustrated in Fig. 2, where we allowed σ_L to take its maximum positive value which occurs when $H_{so} \rightarrow \infty$. The theory in its current state is not adequate to give a quantitative account of the magnetoresistance.

A final point worth noting is that if the parameter β is not set equal to zero but is allowed to act as a free parameter, then Eq. (4) can be made to give a fit as good as the accuracy of the data. This procedure yields a value of H_{so} (~ 1.5 T) which scales with Y concentration but requires a value of β of about 4, which is quite unphysical for Y-Al, so that there is no obvious significance of the fit. We mention it only because it indicates that in superconducting metallic glasses the Maki-Thompson term may mask the deficiencies revealed in nonsuperconducting glasses.

In summary, quantum corrections to the conductivity due to weak localization and Coulombic interaction give at least a semiquantitative description of the low-temperature resistivity and magnetoresistivity of metallic glasses. The temperature dependence of the resistivity is governed predominantly by Coulombic interaction effects and the theory gives a reasonable quantitative account of the results. The magnetoresistance is governed by weak localization, but the theory is only in semiquantitative agreement with the data. It is not clear at present how serious these discrepancies are and whether they could be removed by going to higher order in perturbation theory.

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