Localization, Coulomb interaction, and spin-orbit scattering in amorphous Cu-Ti-Au alloys

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Measurements of the electrical conductivity and magnetoconductivity are reported for a series of Cu-Ti-Au metallic glasses in the temperature range 1.5 to 30 K and in fields up to 7 T. We have found that the theories of weak localization and Coulomb interaction give reasonable agreement with experiment in the strong spin-orbit cases (3 at. % Au and 6 at. % Au), but fails to describe moderate scattering (0.5 at. % Au and 1 at. % Au) results.

INTRODUCTION

The now familiar theories of weak localization and Coulomb interaction have enjoyed considerable success in explaining the anomalous behavior of the transport properties of two-dimensional materials at low temperatures. Recently these theories have also been applied to three-dimensional alloys and a number of papers[1-3] demonstrate reasonable agreement between theory and experiment. The theory of weak localization describes quantum interference of scattered partial waves at defects in the system. Constructive interference of the partial waves leads to enhanced scattering in the backwards direction reducing the conductivity.

Bergmann[4] has recently described the calculation of the Hartree contribution of the Coulomb anomaly in terms of interference giving rise to what he calls "charge holograms." The exchange energy of the electron-electron interaction can be viewed similarly. It opposes the Hartree term, however, which increases conductivity through its effect on the diffusion coefficient.

These theories have been extended to include many effects: spin-orbit scattering,[5] magnetic impurity scattering,[6] and Zeeman spin splitting,[7] as well as orbital magnetic field dependencies.[8] It has been shown[9] that these theories give a good explanation of the change in conductivity as a function of temperature due to increasing spin-orbit scattering with increasing Cu content in Cu-Ti alloys.

The magnetoresistance is also an excellent probe for determining scattering rates and these effects have been investigated in the same series of Cu-Ti alloys.[10] In this paper we shall extend this work to a further manipulation of the strength of spin-orbit scattering by introducing various amounts of Au (0.5 at. %, 1 at. %, 3 at. %, 6 at. %) in $Cu_{35}Ti_{65}$. We will present results for the temperature and magnetic field dependence in a detailed analysis based on the above theories.

EXPERIMENTAL DETAILS

The alloys were prepared by melt spinning arc melted amounts of Cu (99.999% purity), Ti (99.98% purity), and Au (99.999% purity) and were shown to be amorphous by x-ray diffraction. We measured the electrical resistance by a standard four-probe dc method accurate to one part in 10^5 in the temperature range 1.5–30 K. The magnetic field dependence to 7 T was measured at the temperatures 1.5, 5, 7.5, 10, 12.5, 15, and 20 K.

THEORETICAL BACKGROUND

Magnetic field dependence

The magnetoconductance is dominated by the localization effect which, including spin-orbit scattering, Maki-Thompson fluctuations, Zeeman effects, and orbital effects, is given by[5]

$$\Delta\sigma(B,T)=A\left\{B\{F_3(h/(1+t))-\beta F_3(h/t)+C[F_3(h/t_+)-F_3(h/t_-)]\}+\left[\frac{1}{D\tau_{\text{s.o.}}}\right]^{1/2}\left[\frac{-1}{(1-\gamma)^{1/2}}(t_-^{1/2}-t_+^{1/2})+\sqrt{t}-(t+1)^{1/2}\right]\right\}, \tag{1}$$

with

$$A=\frac{e^2}{2\pi^2\hbar},\quad B=\left[\frac{eB}{\hbar}\right]^{1/2},\quad C=\frac{0.5}{(1-\gamma)^{1/2}},$$

$$h=\frac{DeB\tau_{\text{s.o.}}}{\hbar},\quad \gamma=\left[\frac{g\mu_b B\tau_{\text{s.o.}}}{2\hbar}\right]^2,\quad t=\frac{\tau_{\text{s.o.}}}{4\tau_i},$$

$$t_\pm=t+0.5[1\pm(1-\gamma)^{1/2}],$$

$$F_3(z)=\sum_{n=0}^{\infty}\left\{2\left[\left[n+\frac{1}{z}+1\right]^{1/2}-\left[n+\frac{1}{z}\right]^{1/2}\right]-\left[n+\frac{1}{z}+\frac{1}{2}\right]^{-1/2}\right\}.$$

Here D is the diffusion coefficient; $\tau_{s.o.}$ and τ_i are the spin-orbit and inelastic scattering times, respectively. The term $(1-\gamma)^{1/2}$ can in fact become imaginary for the fields and the relaxation times we expect. We have evaluated (1) by treating all variables containing γ as complex; one can, however, easily show that $\Delta\sigma$ remains real. The Zeeman spin effects are contained in γ; setting $\gamma=0$ reduces (1) to the expression of Altschuler and Aronov:[6]

$$\Delta\sigma(B)=\frac{-e^2}{4\pi^2\hbar}(eB/\hbar)^{1/2}[(1+2\beta)F_3(B/B_i) - 3F_3(B/B_{s.o.})] , \quad (2)$$

with

$$B_i=\hbar(\tau_i^{-1}+2\tau_s^{-1})/4eD ,$$

$$B_{s.o.}=\hbar(\tau_i^{-1}+2\tau_{s.o.}^{-1})/4eD .$$

In Eqs. (1) and (2) the function $\beta(T,B)$ is the Maki-Thompson magnetoconductance due to superconducting fluctuations.[11] The exact field dependence of β is not known but it has been suggested[12,13] that the field dependence can be approximated by replacing the temperature-dependent coupling parameter $g(T)=-\ln(T/T_c)^{-1}$, on which β depends, by

$$g(T,B)=-\left[\ln\left[\frac{T}{T_c}\right]+\psi\left[\frac{1}{2}+\frac{DeB}{2\pi k_B T}\right]-\psi(\tfrac{1}{2})\right]^{-1} ,$$

where ψ is the digamma function.

There are two contributions to the magnetoconductivity from Coulomb interactions: Zeeman spin splitting and orbital effects. The spin-splitting term has been calculated by Lee and Ramakrishnan:[14]

$$\Delta\sigma(B)=\frac{-e^2}{4\pi^2\hbar}F_\sigma\left[\frac{k_B T}{2\hbar D}\right]^{1/2}G(B) , \quad (3)$$

where

$$F_\sigma=-\tfrac{32}{3}[1+3F/4-(1+F/2)^{3/2}]F .$$

The term F is the screening factor (see below) and $G(B)$ has been evaluated by Oussett *et al.*[15]

The Zeeman terms are reduced by spin-orbit scattering because of mixing of the spin subbands. Equation (3) will become applicable when $g\mu_b B \gg \hbar/\tau_{s.o.}$, i.e., when $\tau_{s.o.} \geq 10^{-11}$ s. This could certainly be true for the 0.5 at. % and 1 at. % Au alloys. Isawa and Fukuyama[8] have calculated the orbital term

$$\Delta\sigma(B)=\frac{-3}{8}\frac{e^2}{\hbar}\left[\frac{1}{\hbar D}\right]^{1/2}\frac{(k_B T^2)}{(4DeB)^{3/2}}g(T,B)\Phi(T,B) .$$

We have calculated that this term contributes $\simeq 1\%$ to the total magnetoconductivity at the highest field and so has been neglected.

Temperature dependence

Fukuyama and Hoshino[5] have calculated the temperature dependent localization correction to the conductivity and Altschuler and Aronov[6] have modified it to include magnetic scattering τ_s:

$$\Delta\sigma(T)=\frac{e^2}{2\pi^2\hbar D^{1/2}}\left[3\left[\frac{1}{\tau_s}+\frac{1}{\tau_{s.o.}}+\frac{1}{4\tau_i}\right]^{1/2} - \left[\frac{1}{4\tau_i}+\frac{1}{4\tau_s}\right]^{1/2}\right] , \quad (4)$$

where $\tau_{s.o.}$, τ_i, and D are the spin-orbit and inelastic relaxation times and diffusion coefficient, respectively. Only τ_i is temperature dependent and is assumed to be dominated by electron-phonon scattering. In previous papers we have taken $\tau_i \propto T^{-p}$ with $p=2$. However, there is considerable disparity between authors' choice of p—varying between 2 and 3; simple power-law dependences can, however, only be expected to hold in high or low temperature limits. We have also observed quite complicated behavior of τ_i^{-1} crossing over from $p=4$ to $p=2$ over a range from 4 to 20.[10]

Therefore we have decided to use the explicit form for τ_i^{-1} given by Schmid[16] fitting it to the magnetic field results in order that we may remove a parameter from the temperature-dependent analysis. The phase-breaking rate due to inelastic electron-phonon collisions is given by

$$\tau_{i,\alpha}^{-1}=\frac{\pi\hbar^2}{6mMc_\alpha}\int_0^{k_D} dk\, k^2\Phi_\alpha(kl)[\sinh(\hbar c_\alpha k/k_B T)]^{1/2} , \quad (5)$$

where $\alpha=L$ for longitudinal or T for transverse modes; c_α equals sound velocity; l equals elastic mean free path; M equals ionic mass; and k_D equals the Debye wave number. The function Φ can be obtained from Ref. 16.

The limiting behavior of (5) is determined by comparing $k_B T$ and $\hbar c_\alpha/l$. For these alloys we must use the full expression as $\hbar c_\alpha/l \simeq k_B T$.

For sufficiently strong spin-orbit scattering Eq. (4) becomes negative changing localization into antilocalization leading to significant increases in the total conductivity at low temperatures.[17] Magnetic impurities will break the phase coherence destroying the localization effect and increasing the conductivity at low temperatures.[17]

The Coulomb interaction term takes the form:[6]

$$\Delta\sigma(T)=\frac{1.3e^2}{4\pi^2\hbar}\left[\frac{4}{3}-3F^*-\frac{2}{\ln(T_c/T)}\right]\left[\frac{k_B T}{2\hbar D}\right]^{1/2} . \quad (6)$$

Here T_c is the superconducting transition temperature and D is the diffusion coefficient. $F^*=F-\lambda$, where F is an average over the Fermi sphere of the screened Coulomb potential. For Thomas-Fermi screening $F=(1/x)\ln(1+x)$ with $x=(2k_f/k_0)^2$. We can see that F must lie between 0 and 1 and as $k_0^2=4\pi e^2 g(E_f)$, the screening should be quite good ($F \geq 0.4$, the value for a simple metal) and F should vary little between the samples. The term λ is the electron-phonon coupling constant and can be estimated from the McMillan formula.[18]

$Cu_{35}Ti_{65}$ has a T_c of about 60 mK which we have assumed will not change by adding small amounts of gold, so we have kept this fixed for all alloys and thus determined λ to be about 0.3. We note that Eq. (6) does not include the effects of spin-orbit scattering. Fukuyama[19]

has calculated the effect for two dimensions where the result was a reduction in the number multiplying F^* by $\frac{1}{4}$. We do not know the result for three dimensions but note that such a reduction would increase the estimate of F obtained from the fitted results. It is also now believed that the Hartree term will be affected by inelastic scattering so (6) ought to be modified to include τ_i.

ANALYSIS AND RESULTS

Magnetic field dependence

The method of nonlinear least squares was used to fit Eq. (1) to the data. There are two fitting parameters, $\tau_{s.o.}$ and τ_i, for this equation. The diffusion coefficient D, in the original paper,[9] was calculated from the Einstein relation $\sigma(0)=e^2Dg(E_f)$, where the density of states was obtained from specific heat measurements. We have estimated D for the gold alloys by using the Friedel model[20] for the density of states in a transition metal, [because $g(E_f)$ should be dominated by the density of Ti d states] and the measured resistivities. It has been shown[21] that the d states of a transition-metal alloy can account for up to 80% of the conductivity. It is still possible to apply the quantum interference theories to alloys with more than one type of carrier because the parameters appearing in the theory become suitable averages over the Fermi sphere and the number densities of the different carriers.[22] We also note that the interference theories are only applicable in the limit $k_Fl \gg 1$. For our materials k_Fl is about 5 indicating that k_F is no longer a well-defined quantity. However, it has recently been shown[23] that including higher-order terms in $(k_Fl)^{-2}$ in the perturbation expansion will only have a significant effect when k_Fl is very close to 1.

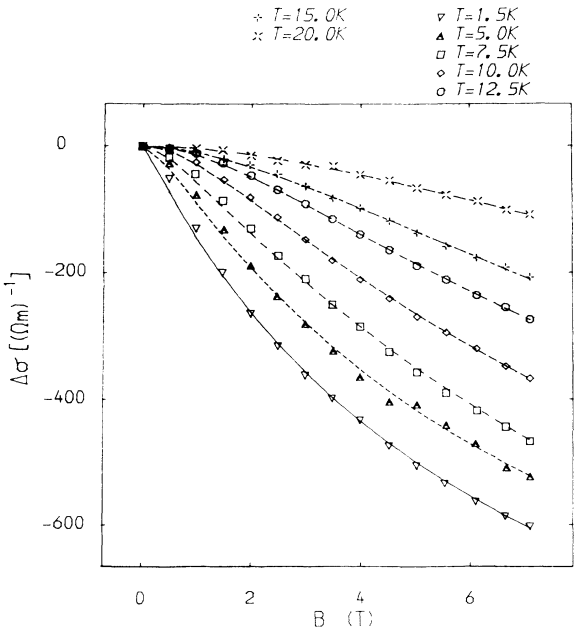

FIG. 1. The change in conductivity $[\Delta\sigma(B)-\Delta\sigma(0)]$ as a function of magnetic field at various temperatures for the alloy $Cu_{35}Ti_{62}Au_3$. The lines are the least-squares best fits.

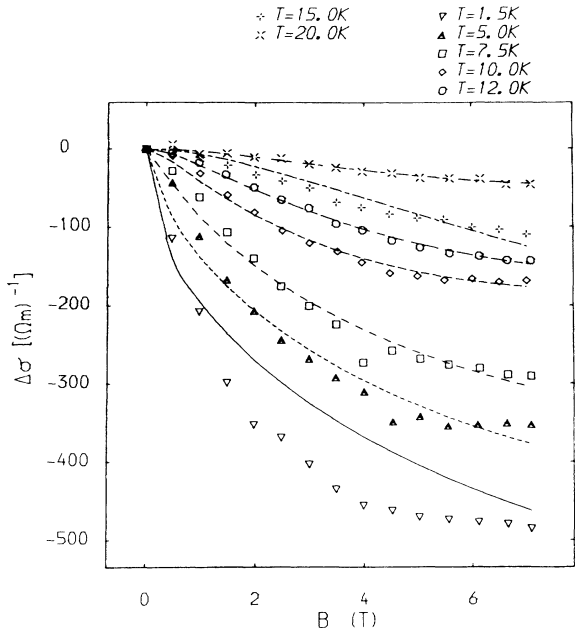

FIG. 2. The change in conductivity $[\Delta\sigma(B)-\Delta\sigma(0)]$ as a function of magnetic field at various temperatures for the alloy $Cu_{35}Ti_{64.5}Au_{0.5}$. The lines are the least-squares best fits.

We have found that (1) will fit only the strongest spin-orbit scattering alloys: the 3 at. % and 6 at. % Au, the remainder return unphysical values of $\tau_{s.o.}$ and very poor fits. As mentioned above the term $(1-\gamma)^{1/2}$ becomes imaginary as the spin-orbit relaxation time increases, so it would appear that although treating the terms as complex seems mathematically acceptable, the theory breaks down as the spin-orbit scattering becomes weaker.

We have also attempted fitting Eq. (2) to the data, but the fits for the 3 at. % and 6 at. % alloys were not nearly as well described. For the 0.5 at. % and 1 at. % results Eq. (2) was much better than (1) although the fits were not good. This is somewhat remedied by including the spin-splitting term (3) though the fits are still unsatisfactory for low temperatures. Figure 1 shows the results for the 3 at. % alloy using (1). The spin-splitting term (3) is not included as this will be greatly reduced in the presence of strong spin-orbit scattering. Figure 2 is the result for the 0.5 at. % sample using both (2) and (3). The relevant parameters are given in Table I where the errors in $\tau_{s.o.}$ are the standard deviations for temperatures at which the fits were reasonable. A peculiar feature of the fits for Eq. (1) was a slight temperature dependence of $\tau_{s.o.}$ tending to decreasing $\tau_{s.o.}$ for increasing temperature. There was no obvious way to avoid this in the fitting procedure.

Of special interest is the temperature dependence of τ_i

TABLE I. Values of the resistivity (ρ), the diffusion coefficient (D), the spin-orbit lifetime ($\tau_{s.o.}$) and the screening parameter (F). The errors in $\tau_{s.o.}$ are the standard deviations for $\tau_{s.o.}$ deduced from the magnetic field analysis at different temperature.

Au conc. (at. %)	ρ ($\mu\Omega$ cm)	D (cm^2 s^{-1})	$\tau_{s.o.}$(10^{-13} s) Temperature dependence	$\tau_{s.o.}$(10^{-13} s) Magnetic field dependence	F
0	182	0.24	73.0	60.0	0.40
0.5	213	0.21	65.6	49.0±6.9	0.42
1	202	0.22	55.0	14.6±4.6	0.38
3	201	0.23	1.8	11.7±8.7	0.38
6	205	0.23	1.2	8.2±4.8	0.41

which is shown in Fig. 3. The solid curve is the result of Schmid (5) where we have used 10 Å for the mean free path, $c_L = 5000$ m s^{-1} for the longitudinal velocity, and $c_T = 1500$ m s^{-1} for the transverse modes.

These estimates are reasonable compared to $c_L = 5205$ m s^{-1} and $c_T = 2434$ m s^{-1} for $Cu_{50}Ti_{50}$ derived from bulk and shear moduli reported in Ref. 24. We will use this expression for τ_i in the temperature dependence below. At low temperatures τ_i becomes temperature independent (for the good fits). This is probably due to magnetic impurities and leads to a magnetic scattering rate τ_s^{-1} of about 2×10^9 s^{-1} from the graph.

Temperature dependence

We have used the same fitting routine for Eqs. (4) and (6) with $\tau_{s.o.}$ and F as fitting parameters. The values of τ_s and τ_i are those determined by the magnetic field analysis as described above. Figure 4 shows the results and the relevant parameters are included in Table I. Agreement between the temperature and magnetic field analyses for $\tau_{s.o.}$ is poor, but the dependence on composition is at least in the right direction. As expected the values of F are quite similar if a little small. This could be due to neglecting the spin-orbit influence on the Coulomb term. It is worth noting that excellent fits to the data can be obtained using $\tau_i \propto T^{-2}$ but the values for F come out negative. Using $\tau_i \propto T^{-3}$, we could fit the data only up to 10 K. Some recently reported values of $\tau_{s.o.}$ are 7×10^{-13} in Y-Al (Ref. 25) and 6×10^{-13} in Cu-Zr (Ref. 26).

In conclusion we have found that the theories of weak localization and electron-electron interaction effects give a semiquantitative description of the temperature and magnetic field dependence of the conductivity in the strong spin-orbit case. Although the two analyses do not agree

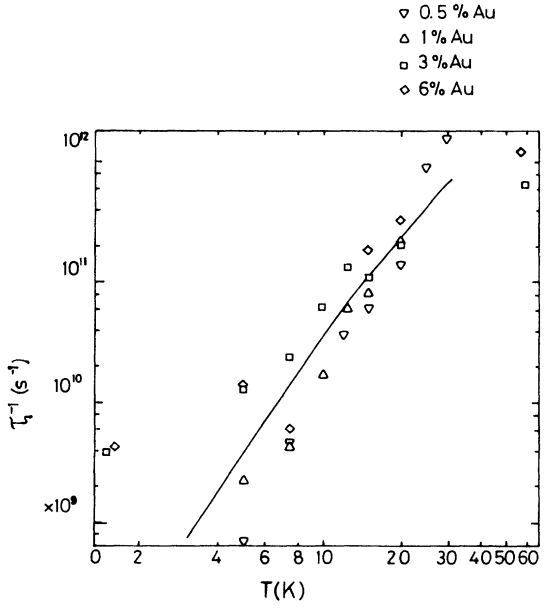

FIG. 3. The temperature dependence of the phonon inelastic scattering rate for various alloys. The solid line is the theoretical expression for τ_i^{-1} [Eq. (5)] fitted using values for the sound velocities.

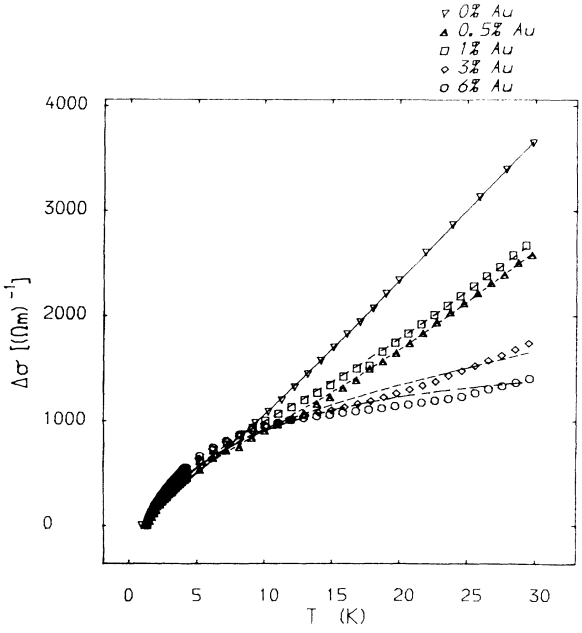

FIG. 4. The change in the conductivity $[\Delta\sigma(T)-\Delta\sigma(1.5)]$ as a function of temperature for various alloys. The lines are the least-squares best fits.

in their estimates of $\tau_{s.o.}$, there is a trend of increasing $\tau_{s.o.}^{-1}$ with increasing Au content. In the magnetic field analysis the variance with temperature of $\tau_{s.o.}$ is quite large and shows a slight temperature dependence. For moderate spin-orbit scattering the agreement is quite poor. It would seem that theories where the spin-orbit effect has been included work reasonably well in the limits of very weak or very strong scattering, but do not describe the intermediate scattering region. Other difficulties in applying these theories have been recently reported, in particular Refs. 25 and 26.

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