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Localization and electron-interaction effects in metallic glasses

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Magnetoresistance measurements have been made on amorphous $Lu_{75}Pd_{25}$ and $Lu_{75}Ni_{25}$ alloys. In the absence of contributions from spin-splitting interaction (because of the high spin-orbit scattering rate $1/\tau_{so} \simeq 10^{14}$ s^{-1}) and superconducting fluctuations, direct comparison of experimental data with three-dimensional weak localization theory can be made. Our results follow the trend predicted by theory, but are about 20% larger. Interaction effects with a negative screening parameter can explain both the discrepancy and the order of magnitude of resistivity $\rho(T)$. We obtain the inelastic scattering time $\tau_i(T) \simeq 2\times10^{-10}T^{-2}$ s which can be attributed to electron-phonon processes in disordered metals, according to the theory of Bergmann and Takayama. Prior results obtained for amorphous $Zr_{40}Cu_{60}$ are discussed only qualitatively as this alloy does exhibit superconducting fluctuations above 0.5 K.

Magnetoresistance measurements in thin films have led to the determination of parameters for the inelastic and spin-orbit scattering processes governing the transport properties in disordered metals.[1] Recently, similar studies have been extended to three-dimensional (3D) metallic systems including granular aluminum[2] and metallic glasses.[3] In these systems, four contributions to magnetoresistance are present: localization,[4] two types of electron-electron interactions because of the orbital effect[4] and spin-splitting effect,[5] and superconducting fluctuations.[4,6] Methodically, it is quite difficult to separate these contributions in order to extract the physical parameters. Ideally, the procedure will be greatly simplified if some of the interaction effects can be suppressed. Then, a direct comparison with localization theory can be made. In this Rapid Communication, we report magnetoresistance studies in amorphous Lu-based alloys (high atomic number). These materials were chosen because of the high spin-orbit scattering (spin-mixing) rate and absence of superconductivity in them. Thus, the spin-splitting interaction and superconducting fluctuations can be neglected. Also, because of strong spin-orbit interaction, the orbital contribution is reduced to one-fourth of its normal value.[4] Hence, one can focus mainly on the localization effect.

Amorphous $Lu_{75}Pd_{25}$ and $Lu_{75}Ni_{25}$ samples were prepared in ribbon form by melt spinning. Amorphicity of the samples were checked by x-ray diffraction. Magnetoresistance measurements were made in longitudinal fields of up to 85 kOe and temperatures down to $\sim$0.7 K. All measurements were carried out in a four-point probe. Resistivity as a function of temperature $\rho(T)$ for $Lu_{75}Pd_{25}$ is shown in Fig. 1 (inset). We did not observe any precursor to superconducting fluctuation down to $\sim$0.5 K. Similar results were observed in $Lu_{75}Ni_{25}$. Measurements were also made on thermally relaxed samples.[7] The results were the same. Different from amorphous $Zr_{40}Cu_{60}$ alloys studied earlier,[8] $\rho(T)$ does not strictly follow a square-root variation with temperature, as indicated in Fig. 1 (inset). This will be discussed with respect to strong spin-orbit scattering. Magnetoresistance data for $Lu_{75}Pd_{25}$ at six different temperatures are plotted in Fig. 1. Results on $Lu_{75}Ni_{25}$ are almost identical.

The two contributions to magnetoresistance in our samples need to be considered are as follows.

(a) Localization incorporating Zeeman spin-splitting and spin-orbit interaction derived by Fukuyama and Hoshino[9]:

$$\frac{\Delta\rho}{A\rho^2} = -\sqrt{h}\,f_3\left(\frac{1+t}{h}\right) - 0.5\left(\frac{h}{1-\gamma}\right)^{1/2}\left[f_3\left(\frac{t_+}{h}\right) - f_3\left(\frac{t_-}{h}\right)\right] + \frac{1}{\sqrt{1-\gamma}}(\sqrt{t_-} - \sqrt{t_+}) + \sqrt{t+1} - \sqrt{t}\ , \quad (1)$$

where

$$A = \frac{e^2}{2\pi^2\hbar\sqrt{D\tau_{so}}} = \frac{1.23\times10^{-5}}{\sqrt{D\tau_{so}}}\ , \quad (2a)$$

$$h = \frac{eDH\tau_{so}}{c\hbar} = 1.518\times10^{10}D\tau_{so}H \quad (H \text{ in kOe})\ , \quad (2b)$$

$$\gamma = \left(\frac{0.58}{D}\right)^2 h^2\ , \quad (2c)$$

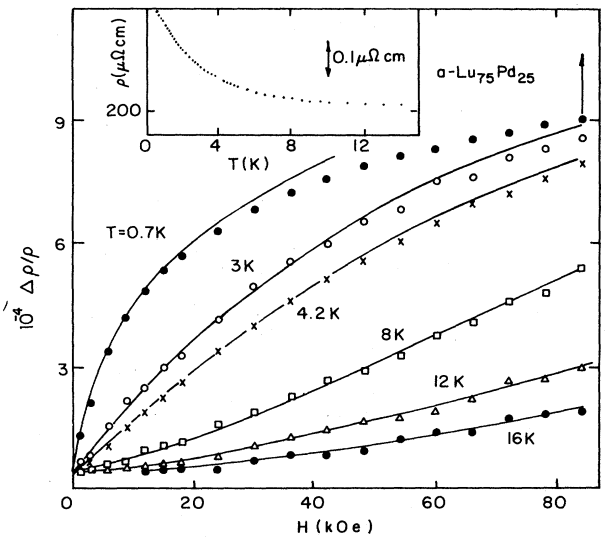

FIG. 1. Normalized magnetoresistivity as a function of applied field at various temperatures for amorphous $Lu_{75}Pd_{25}$. Results on $Lu_{75}Ni_{25}$ are similar. Solid curves are fits to the localization theory of Fukuyama and Hoshino ($D = 1.2$ cm^2/s). Vertical arrow indicates deviation due to orbital-interaction effect. The theoretical curves have been scaled by a factor of 1.2 (see text). Inset: resistivity as a function of temperature for $Lu_{75}Pd_{25}$.

$f_3(x)$ is the Kawabata function[10] for 3D disordered system, D is the electronic diffusivity, $t=\tau_{so}/4\tau_i$, τ_i is the inelastic scattering time, $t_\pm = t+0.5(1\pm\sqrt{1-\gamma})$. For $\gamma>1$, the modification of expression (1) is straightforward. Numerical results for several values of t with $D\simeq 1.2$ cm^2/s are illustrated in Fig. 2.

(b) Electron interaction in the particle-particle channel[4]:

$$\frac{\Delta\rho}{\rho^2}=\frac{1}{4}\left(\frac{e^2}{2\pi^2\hbar}\right)\left(\frac{eH}{\hbar c}\right)^{1/2} g(T,H)\Phi_3(y) \quad . \tag{3}$$

The function $\Phi_3(y)$ is given in Ref. 5 with $y=2DeH/\pi ck_BT$. The factor $\frac{1}{4}$ is included for strong spin-orbit scattering. g is the interaction parameter in the "particle-particle channel,"[4] $g<0$ for attractive interaction and $g>0$ for repulsive interaction leading to negative and positive magnetoresistance, respectively.

A rough estimate of the contribution from orbital effect is made by taking the diffusivity $D\sim 1$ cm^2/s and $g\sim 0.2$ for normal metals. The result account for less than 4% of the total magnetoresistance at $H=85$ kOe and above ~ 3 K. In fact, the experimental data (with exceptions for $H>30$ kOe at $T=0.7$ K) shown in Fig. 1 follow the trend of the theoretical curves depicted in Fig. 2. Thus, a first-order comparison with localization theory can be made to determine τ_{so} and $\tau_i(T)$ provided D is known. The simplicity of this procedure (with only two fitting parameters) leads to a rather unambiguous determination of the inelastic scattering rate and a better understanding of the scattering processes involved. The electron diffusivity can be determined from conductivity $\sigma=e^2N(0)D$, where $N(0)$ is the total density of states at the Fermi level. For $Lu_{75}Pd_{25}$, $N(0)$ is estimated to be ~ 0.6 states/eV atom using information on the s and d band occupancies in Lu (Ref. 11) and the shape of the density of states in liquid La.[12] We obtain $D\simeq 1.2$ cm^2/s for $Lu_{75}Pd_{25}$. The data were fitted to expression (1) for $H\lesssim 30$ kOe at $T=0.7$ K and for $H\lesssim 60$ kOe for $T\gtrsim 3$ K. An overall good fit is obtained if one increases the theoretically predicted strength of localization by $\sim 20\%$. This discrepancy will be discussed. The results are shown by the solid lines in Fig. 1. We obtain from curve fitting the spin-orbit scattering time $\tau_{so}\simeq 3\times 10^{-14}$ s and the inelastic lifetime of conduction electrons $\tau_i\simeq 1.9\times 10^{-10}T^{-2}$ s.

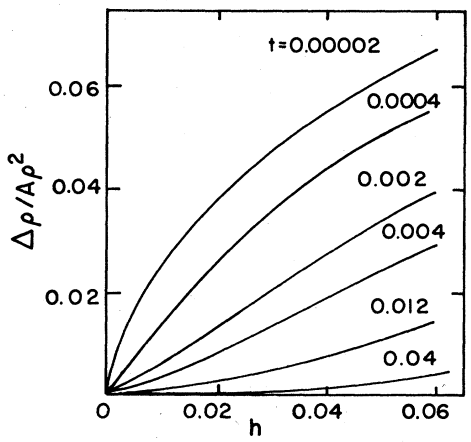

FIG. 2. $\Delta\rho/A\rho^2$ vs h for different values of t obtained from Eqs. (1) and (2). D is chosen to be 1.2 cm^2/s.

The results for $\tau_i(T)$ are shown in Fig. 3. Vertical bars denote uncertainties in our curve-fitting procedure. It is noted that at low temperature (0.7 K), the data are insensitive to the choice of t and thus τ_i.

Let us now comment on the orders of magnitude of the physical parameters τ_{so} and τ_i. The spin-orbit scattering time τ_{so} in some metallic glasses can be determined from critical-field results. It was shown that the value of τ_{so} obtained compared well with the trend predicted from atomic calculations.[13,14] For nonsuperconducting Lu-based glasses, one can then estimate using Ref. 14 that $\tau_{so}\sim 10^{-14}$ s, in good agreement with our present finding. The lifetime of conduction electrons is limited by electron-electron and electron-phonon scatterings. For the former process, Schmid[15] showed that for impure metals, the inverse lifetime is given by

$$\frac{\hbar}{\tau_{ee}}=\frac{\pi E^2}{8E_f}+(\sqrt{3/2})(1/k_Fl)^{3/2}(E^{3/2}/E_F^{1/2}) \quad ,$$

where $E=k_BT$, E_F is the Fermi energy, k_F is the Fermi wave number, and l is the electronic mean free path. Bergmann[16] and Takayama[17] derived an expression for the electron lifetime because of the electron-phonon scattering in impure metals. The inverse lifetime $(\hbar/\tau_{e\text{-ph}})=(2\pi^2\alpha/k_Fl)(k_BT)^2/\hbar\omega_D$, where the coupling constant $\alpha\simeq 1$ and ω_D is the Debye frequency. To compare with experimental results in $Lu_{75}Pd_{25}$, we take for the Debye temperature $\theta_D\simeq 140$ K,[13] $E_F\simeq 1$ eV, and $k_Fl\simeq 2$ (free-electron results). This yields

$$1/\tau_{e\text{-ph}}\simeq 10^{10}T^2 \text{ s}^{-1} \quad ,$$

$$1/\tau_{ee}\simeq(4.46\times 10^6T^2+4.2\times 10^8T^{3/2}) \text{ s}^{-1} \quad .$$

Our results indicate that $1/\tau_i\simeq 5.2\times 10^9T^2$ s^{-1}. Thus, the

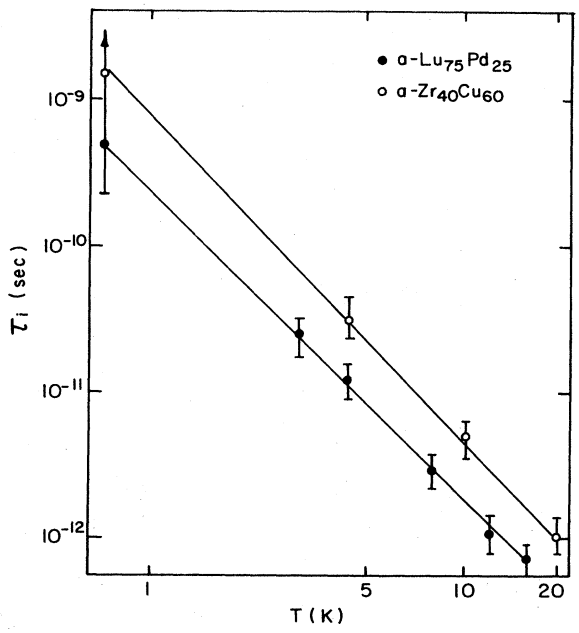

FIG. 3. $\log\tau_1$ vs $\log T$ to determine the temperature dependence of the electronic lifetimes for amorphous $Lu_{75}Pd_{25}$ and $Zr_{40}Cu_{60}$ alloys. Data are derived from Fig. 1 and results in Ref. 8. Vertical bars denote uncertainties in the curve-fitting procedure.

electron scattering processes in our alloys are clearly dominated by electron-phonon scattering for temperatures above ~ 0.7 K.

To account for the deviations observed at high fields (~ 85 kOe) and in low temperatures (~ 0.7 K) in Fig. 1, the orbital effect on localization needs to be considered. Comparing our data to the theoretical curves given in Fig. 2 suggests that the interaction is attractive. At $T=0.7$ K, we obtain $g \simeq -0.4$ using expression (3) and data shown in Fig. 1 (vertical arrow). For attractive interaction mediated by electron-phonon interaction, $g^{-1} \simeq 1/\lambda + \ln(\theta_D/\theta)$, where $\theta = \max(k_B T, DeH/c)$. This gives $\lambda \simeq -0.13$. The electron-phonon interaction parameter $\lambda_{e\text{-ph}}$ is given by $\lambda - \mu^*$. Taking the Coulomb pseudopotential μ^* to be 0.13, one obtains $\lambda_{e\text{-ph}} \simeq 0.26$. This low value of $|\lambda_{e\text{-ph}}|$ accounts for the absence of superconductivity in the Lu alloys. The temperature-field dependence of g and Φ_3 [Eq. (3)] is also found to account for the reduced deviations above 0.7 K in high fields.

Earlier, attempts were made to separate the four contributions to magnetoresistance in amorphous $Zr_{40}Cu_{60}$ (with superconducting transition temperature T_c at ~ 0.2 K) alloys.[8] It was noted that good fits to localization theory[9] could only be obtained if a rather long spin-orbit scattering time $\tau_{so} \simeq 2.5 \times 10^{-12}$ s was introduced. The latter is an order of magnitude longer than that expected for the $4d$ metals.[13] It should be noted, however, that the spin-orbit parameters λ_{so} for the Zr-Cu alloys[18] determined from critical fields are ~ 2. This suggests that $\tau_{so}(\alpha 1/\lambda_{so} T_c)$ for $Zr_{40}Cu_{60}$ ($T_c \simeq 0.2$ K) should be a factor of 10 higher than those observed in Zr alloys ($T_c \simeq 4$ K).[13] For $Zr_{40}Cu_{60}$, however, only qualitative comparison with localization theory is possible since a quantitative description of electron interaction (b) and superconducting fluctuations in high fields is still lacking.[2] Discrepancy between experimental results and that predicted from localization theory is about 10–20%. Using results from Ref. 8, one obtains $1/\tau_i \simeq 10^9 T^{2.2}$ s^{-1} as shown in Fig. 3. This is in qualitative agreement with results based on electron-phonon interaction.

Returning to the question of discrepancy ($\sim 20\%$) between experimental results and theoretical predictions, we can only make the following conjecture. It was recalled that in n-InSb, the strength of the observed localization effect was half of that expected from theory.[19] This reduction in strength was attributed to electron-electron correlation effects characterized by a Coulomb screening parameter F (varies from 0 to 1 in the conventional theory). On the other hand, a negative F might have the opposite effect. Recently, negative values of F were reported for granular Al.[2] Implications of the latter and its effects on localization await further theoretical investigation. We also find it necessary to have $F < 0$ for explaining the order of magnitude of $\rho(T)$ in our Lu alloys. Combining electron-electron interaction[5] and localization[9] effects on resistivity in zero field, the variation in magnitude of the latter between 0.5 and 4 K can only be explained if $F \simeq -0.6$. The departure from $T^{1/2}$ behavior in the $Lu_{75}Pd_{25}$ is due to strong spin-orbit scattering. On the other hand, $\rho(T)$ in $Zr_{40}Cu_{60}$ (weak spin-orbit scattering) varies as $T^{1/2}$ up to ~ 9 K. Using data from Ref. 8, a value of $F \simeq -0.05$ is obtained by fitting $\rho(T)$ to the electron-electron interaction theory.[5] It is also emphasized that we have used the measured values of the electron diffusivity [derived from σ and $N(0)$] in our analysis.

In summary, we have performed magnetoresistance measurements on amorphous Lu-based alloys. The simplicity of the system permits a direct comparison with localization theory. It is shown that the transport properties in metallic glasses can be described by weak localization ($k_F l > 1$). Values of the spin-orbit scattering rates obtained from different measurements are in good agreement. Inelastic scattering are dominated by electron-phonon processes at temperatures above ~ 1 K. A discrepancy of $\sim 20\%$ is observed which is attributed to attractive electron-electron correlation. The negativity of the screening parameter F is not consistent with the present status of the theory.

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