Quantum interference effects in $(Ni_{0.5}Zr_{0.5})_{1-x}Al_x$ metallic glasses

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(Received 22 August 1996)

Very high-resolution electrical resistivity data are presented here for $(Ni_{0.5}Zr_{0.5})_{1-x}Al_x$ (with $x=0$, 0.05, 0.1, 0.15, 0.2) nonmagnetic amorphous metallic glasses in the temperature range 1.2–300 K. All the alloys fall into the strong scattering regime with negative temperature coefficient of resistance even until 300 K. *In each specimen*, we find three distinct regions, hitherto only theoretically predicted by quantum interference effects, where the conductivity varies in a sequence of $\sqrt{T}$, T, and $\sqrt{T}$ at low, intermediate, and high temperatures, respectively. Parameters estimated from this quantum correction analysis are in excellent agreement with those from earlier experiments on other amorphous systems. [S0163-1829(97)06310-8]

In strongly disordered amorphous materials the occurrence of negative temperature coefficient of resistivity (TCR $\sim\rho^{-1}d\rho/dT$), giving rise to resistivity minima[1,2] at low temperatures, have stimulated many investigators to explore the various scattering mechanisms. The increase of $\rho(T)$ below $T_{\min}$ was initially attributed to scattering from the structural two-level states $[\approx -\ln(T^2+\Delta^2)]$ or to a magnetic one of the Kondo type ($\approx -\ln T$). Rapp *et al.*[3] have reanalyzed the data for many metallic glass systems, previously fitted to the $-\ln T$ relation, and have concluded that $-\sqrt{T}$ fits much better below $T_{\min}$. More recent theories,[4,5] considering quantum corrections to the Boltzmann conductivity equations, namely, weak localization, and electron-electron interaction effects, offer an interpretation to the negative TCR in metallic glasses.[6,7]

The electrical transport properties of amorphous transition-metal alloys have been well reviewed by Mizutani,[1] Howson and Gallagher,[7] and Dugdale.[8] Among them, nonmagnetic highly resistive metallic glasses[1] in which the magnetic susceptibility is negligibly small and temperature independent (Pauli paramagnet with $\chi \sim 10^{-5} - 10^{-6}$ emu/mol), serve as the most suitable system to study the motion of conduction electrons in highly disordered environments. Some of these metallic glasses are[1,9] Cu-Ti, Cu-Zr, Ni-Zr, Ni-Zr-M (M=Al, B, Si, H), Ni-P, etc. Generally they fall into the strong scattering regime ($\rho_{300K}>150\mu\Omega$ cm) with negative TCR and typical electron mean free path (l) of the order of average interatomic spacing ($\sim$ 3–5 Å).

When the electron mean free path becomes comparable with the interatomic distance an incipient weak localization along with an enhanced electron-electron interaction show up. The electron-electron interaction effect (EEI) arises from the modified Coulomb interaction between electrons which are weakly localized due to elastic impurity scattering. Two theoretical approaches exist which predict a low-temperature $\sqrt{T}$ variation of conductivity (σ). In the strong-scattering limit, McMillan's[2] scaling theory of metal-insulator transition yields for the conductivity on the metallic side, $\sigma(T)=\sigma(0)[1+(T/\Delta)^{1/2}]$, where Δ is the correlation gap in the density of states at E_F. In the perturbative interaction approach,[4,5] the temperature dependence of the conductivity $\sigma(T)$, valid for three-dimensional weak-scattering materials, is given by[4,5]

$$\sigma(T)-\sigma(0)=\frac{1.3}{\sqrt{2}}\frac{e^2}{4\pi\hbar}\left(\frac{4}{3}-\frac{3}{2}F_\sigma\right)\sqrt{\frac{k_BT}{\hbar D}}, \qquad (1)$$

where D is the diffusion constant, and F_σ is the screening factor for the Coulomb interaction. Cochrane and Strom-Olsen[10] deduced a quasiuniversal temperature variation of conductivity $\sigma(T)-\sigma(0)=\Delta\sigma(T)=(500\pm100)\sqrt{T}(\Omega m)^{-1}$, valid for a large number of high resistivity metallic glasses with a wide range of $\sigma(0)$.

In strongly disordered crystalline and amorphous alloys the electron localization[4] plays a significant role in determining the sign and magnitude of TCR at temperatures even as high as the room temperature. The mutual interference between waves elastically scattered from nearby ions leads to a phase coherence between the scattered partial waves. As a consequence, the probability for an electron to return to its origin is enhanced which implies a tendency of localization. Quantum considerations[5] increase the probability of backscattering. Inelastic scattering and magnetic field can, however, destroy the phase coherence and reduce the additional resistivity. Lee and Ramakrishnan,[4] Altshuler and Aronov,[5] and Dugdale[8] have given detailed theoretical and physical interpretations of the weak localization and the EEI effects.

Using a scaling approach to localization by Abrahams *et al.*[11] and further simplification by Howson,[12] the correction to the Boltzmann conductivity is given by $\Delta\sigma=e^2/[\pi^2\hbar L_i(T)]$, where L_i is the inelastic diffusion length. $L_i^2(T)=1/2l_el_i(T)$, where l_e and l_i are elastic and inelastic mean free paths. This was shown[12] to lead to a temperature dependence in amorphous alloys of the form

$$\Delta\sigma\propto T, \quad \Theta_D/10<T<\Theta_D/3 \qquad (2)$$

and

$$\Delta\sigma\propto\sqrt{T}, \quad \Theta_D/3<T<\Theta_D. \qquad (3)$$

This localization correction to the conductivity [Eqs. (2) and (3)] describes adequately the temperature dependence from 20 to 300 K in a number of nonmagnetic transition-metal-based alloys,[6,7,12] namely, $Cu_{50}Ti_{50}$, $Cu_{50}Zr_{50}$, $Cu_{50}Hf_{50}$, $Ti_{50}Be_{40}Zr_{10}$, etc.

TABLE I. Fit to $\sigma(T)=\sigma_0+m_\sigma\sqrt{T}$ in the temperature range ($T_c \leqslant T \leqslant 15$ K) : Al conc. (x), coefficients $\chi^{2\,a}$, range of fit, diffusion constant (D), and density of states $N(E_F)$ for $(Ni_{0.5}Zr_{0.5})_{1-x}Al_x$ alloys.

x Al conc.	σ_0 $(\Omega\ m)^{-1}$	m_σ $(\Omega\ m)^{-1}K^{-1/2}$	$\chi^{2\,a}$ (10^{-10})	Range of T (K)	D 10^{-5} (m^2/s)	$N(E_F)$ $(atom\ eV)^{-1}$
0.00	530079	358.9	0.2	4–11	5.8	0.95
0.05	480734	365.8	0.5	4–15	5.5	0.88
0.10	457690	379.2	0.7	3–15	5.1	0.89
0.15	443267	438.4	1.0	3–10	3.8	1.18
0.20	441340	458.8	1.0	2–8	3.5	1.20

[a] $\chi^2=1/N\Sigma_{i=1}^{i=N}[Y_i(\text{data})-Y_i(\text{fit})]^2/[Y_i(\text{data})]^2$

All the amorphous samples under investigation are in the form of ribbons having 20–30 μm thickness and 1–1.5 mm width. A conventional four-probe dc technique has been employed to measure the electrical resistivity $[\rho(T)]$ of $(Ni_{0.5}Zr_{0.5})_{1-x}Al_x$ with $x=0$, 0.05, 0.1, 0.15, 0.2 in the temperature range 1.2–300 K. A combination of a temperature controller (Lake Shore, DRC-93C), a current source (Keithley, 220), and a digital multimeter (Keithley, 196) was employed. Liquid He^4 was used as a coolant in a homemade cryostat and the lowest temperature of 1.2 K was attained by pumping on the He^4 bath by a very high speed Kinney pump. Typical measuring current of 15–30 mA has been used to minimize any heating effect in these highly resistive metallic glasses even at 1.2 K. We found that larger measuring currents ($>$ 50 mA) disturb the thermal stability considerably at the lowest temperature of 1.2 K. The current and voltage contacts with the sample were made with a nonsuperconducting Zn-Cd solder. $\rho(T)$ data were taken at intervals of 50 mK in the range of 1.2 to 50 K and then 100 mK till 300 K. The geometrical factors were determined from the mass, density, length, and width. The absolute accuracy of the resistivity is only within 5% due to the uncertainties in the geometrical factors. The relative accuracy, however, was better than 1–5 parts in 10^6. The temperature stability during each measurement was ± 10 mK.

We find that all the samples show negative TCR until the highest temperatures (300 K). However, each of them shows a tendency towards superconductivity below 4 K. The superconducting transition temperature (T_c) and its onset are found to decrease with increasing Al content. The room-temperature values of the resistivity ($\rho_{300\,K}$) for all the alloys are high ($>$ 165 $\mu\Omega$ cm) and they increase with the Al concentration (x). The motivation behind the present work is to check whether the scattering mechanism in all these highly disordered nonmagnetic alloys can be well described by quantum interference effects (QIE). Specifically, whether the theoretical[4,11,12] sequence of $\sqrt{T}$, T, and $\sqrt{T}$ dependence of conductivity could be observed at low-, intermediate-, and high-temperature ranges. Until now, there has been only one report,[2] to our knowledge, on Co-rich ferromagnetic bulk (three-dimensional) metallic glasses where the full sequence was observed in the same alloy. However, since ferromagnetism may destroy localization,[4,8], the interpretation could be doubted. In the present study all the alloys are nonmagnetic having temperature-indepedent Pauli paramagnetism ($\chi\sim 10^{-5}-10^{-6}$ emu/mol).[13]

In the temperature range $T_c<T<15$ K, the data could be fitted very well to Eq. (1), viz., $\sigma(T)=\sigma(0)+m_\sigma\sqrt{T}$. In Table I, we have summarized the coefficients of the fits, the values of χ^2 (quality of fit), and the ranges of fit, the latter gets extended towards lower temperatures as Al concentration (x) suppresses T_c. Figure 1 clearly shows this low-temperature $\sqrt{T}$ dependence of the conductivity which changes by merely 0.1%. The quality of fit ($\chi^2\sim 10^{-10}$) is extremely good and consistent with the high experimental accuracy. The coefficient of $\sqrt{T}$ $[m_\sigma\approx(360–500)\ (\Omega\ m)^{-1}\ K^{-1/2}]$ is in good agreement with the near-universal value[10] of $(500\pm100)\ (\Omega\ m)^{-1}\ K^{-1/2}$ and those obtained by Das and Majumdar[2] on ferromagnetic metallic glasses.

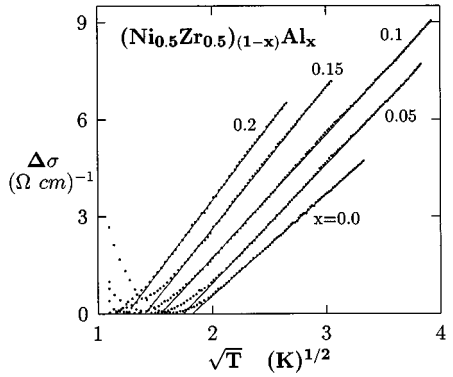

FIG. 1. Plot of $\Delta\sigma$ vs $\sqrt{T}$ for $(Ni_{0.5}Zr_{0.5})_{1-x}Al_x$ alloys and the best-fitted straight lines (Table I) as predicted by the EEI theory.

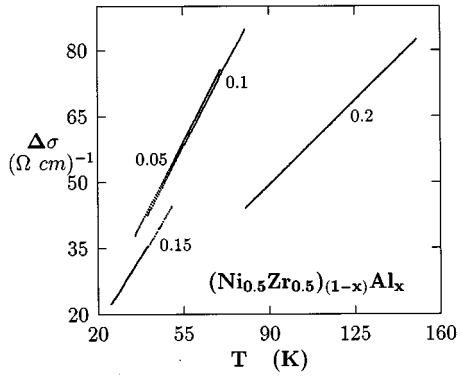

FIG. 2. Linear dependence of $\Delta\sigma$ on T in the intermediate range (Table II) as predicted by the localization theory.

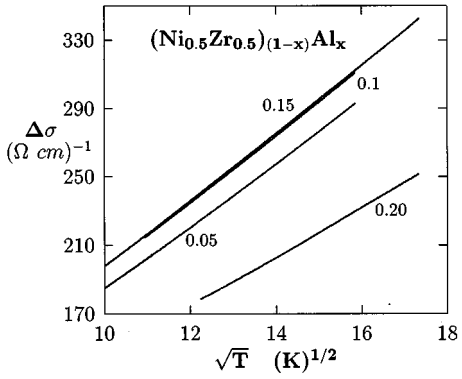

FIG. 3. Linear dependence of $\Delta\sigma$ on $\sqrt{T}$ in the high-temperature range (Table II) as predicted by the localization theory.

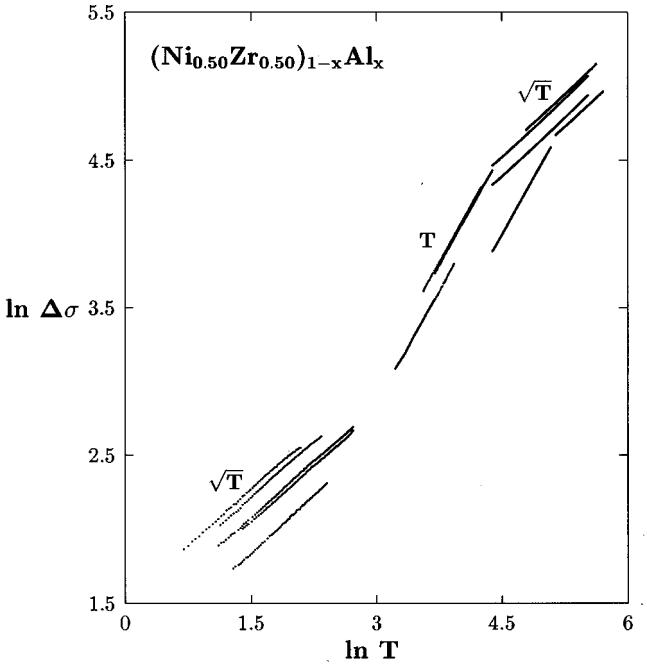

FIG. 4. Plot of $\ln(\Delta\sigma)$ vs $\ln T$ depicting three distinct regions of $\sqrt{T}$, T, and $\sqrt{T}$ dependence in agreement with QIE. Only raw data are shown in Figs. 2–4.

For all these alloys $K_F l \sim 4$–5 (K_F = Fermi wave vector) and so one should expect the screening parameter F_σ in Eq. (1) to be negligibly small. The diffusion constant (D), obtained from the slope (m_σ) of $\sigma(T)$ and Eq. (1) keeping $F_\sigma = 0$, is in the range $(3.5$–$5.8) \times 10^{-5}$ (m^2/s) for all five samples (Table I). It is this D which carries the signature of disorder and we find that $\rho_{300} \propto 1/D$ for our alloys. The values are similar to those obtained by Howson and Greig ($\approx 2.0 \times 10^{-5} m^2/s$ for Cu-Ti, Cu-Zr, and Cu-Hf alloys[6]) and Das and Majumdar[2] [$\approx$ $(0.4$–$4.0) \times 10^{-5}$ m^2/s]. Consequently, the value of the density of states [$N(E_F)$], calculated using the Einstein relation $D=[\rho(0)e^2N(E_F)]^{-1}$, is about 1.0 $(\text{atom ev})^{-1}$ (Table I) for all the five alloys. This value is in excellent agreement with those found in CuZr and NiZr nonmagnetic metallic glasses.[1] All these results lead to the conclusion that our measured slope (m_σ) for the $\sqrt{T}$ dependence of $\sigma(T)$ can be well explained in terms of the electron-electron interaction theory.

Θ_D of all these alloys are in the range of 250 to 350 K. The region roughly between $\Theta_D/10 < T < \Theta_D/3$ clearly shows for all the alloys a linear temperature dependence of $\sigma(T)$ as given by Eq.(2). Figure 2 shows these linear dependences and Table II gives all the relevant parameters. The values of χ^2 are consistent with the experimental error indicating excellent quality of fit. In the region $\Theta_D/3 < T < \Theta_D$, a $\sqrt{T}$ behavior, as given by Eq. (3) is found for all these alloys. Figure 3 clearly shows the $\sqrt{T}$ dependence and Table II gives the relevant parameters. In this regime as well the fits are excellent ($\chi^2 \approx 10^{-10}$). The values of $\sigma(0)$ (which is the extrapolated value of σ at $T=0$ K) are not exactly the same (Tables I and II) for each sample. This kind of behaviour of $\sigma(0)$ was also obtained by Howson and Greig[12] for nonmagnetic transition-metal-based alloys. Moreover, Das

TABLE II. Fit in the temperature ranges $\Theta_D/10 \leq T \leq \Theta_D/3$ and $\Theta_D/3 \leq T \leq \Theta_D$: Al conc.(x), coefficients χ^2, range of fit, inelastic mean free paths (l_i), equations used, and inelastic relaxation times (τ_i) for $(Ni_{0.5}Zr_{0.5})_{1-x}Al_x$ alloys.

x Al Conc.	σ_0 $(\Omega\ m)^{-1}$	m'_σ $(\Omega\ m)^{-1}K^{-1}$ or $(\Omega\ m)^{-1}K^{-1/2}$	χ^2 (10^{-10})	Range of T (K)	l_i $10^{-4}(mT^{-2})$ or $10^{-6}(mT^{-1})$	Fit of σ to	τ_i $(10^{10}\ T^2)^{-1}$ s or $(10^{13}\ T)^{-1}$ s
0.00	529617	112.4	1.7	30–75	3.2	$\sigma_0 + m'_\sigma T$	2.1
	522612	1740.7	8.4	105–250	1.3	$\sigma_0 + m'_\sigma \sqrt{T}$	8.7
0.05	479911	107.6	1.3	35–70	3.5	$\sigma_0 + m'_\sigma T$	2.4
	471749	1837.8	9.1	100–250	1.2	$\sigma_0 + m'_\sigma \sqrt{T}$	8.1
0.10	456949	105.7	8.9	40–80	3.6	$\sigma_0 + m'_\sigma T$	2.6
	447619	1954.3	4.3	120–250	1.1	$\sigma_0 + m'_\sigma \sqrt{T}$	7.7
0.15	443280	87.8	3.7	25–50	5.2	$\sigma_0 + m'_\sigma T$	5.1
	432921	1966.9	4.1	100–300	1.0	$\sigma_0 + m'_\sigma \sqrt{T}$	10.1
0.20	440091	54.8	1.2	80–140	13.4	$\sigma_0 + m'_\sigma T$	14.3
	430475	1447.5	9.4	150–300	1.9	$\sigma_0 + m'_\sigma \sqrt{T}$	20.5

and Majumdar[2] also found different values of $\sigma(0)$ when extrapolated from three distinct temperauture regions in the case of Co-rich metallic glasses. The reason for this is still not clear. Nevertheless, in our case the variation in $\sigma(0)$ does not in any way influence the analysis. However, we should point out that we do find a systematic decrease of $\sigma(0)$ with x (Al. conc.) in each region consistent with the observation of the increase of ρ with x.

In Fig. 4 we have shown separately the three distinct regions of $\sqrt{T}$, T, and $\sqrt{T}$ on a log-log plot. The parallellism of the lines is indeed striking, especially those between the two extreme regions of temperature where $\Delta\sigma$ has the same functional dependence.

Taking the elastic mean free path length (l_e) $\sim$ 3 Å which is of the order of interatomic spacing for all our alloys,[9] we obtain (shown in Table II) from the slope of the $\sigma(T)$ curve the inelastic mean free path length to be $l_i(T)=(3.2-13.4)\times10^{-4}T^{-2}$ (m) for the Ni-Zr-Al alloys in the temperature range $\Theta_D/10<T<\Theta_D/3$. In the region $\Theta_D/3<T<\Theta_D$, the same $l_i(T)=(1.0\text{–}1.9)\times10^{-6}\ \mathrm{T}^{-1}$ (m). All these values are very similar to those of $7.7\times10^{-4}\ \mathrm{T}^{-2}$(m) for $T\leqslant\Theta_D/3$ and $1.8\times\ 10^{-6}T^{-1}$ (m) for $T>\Theta_D/3$ reported in the case of nonmagnetic transition- metal based alloys[12].

The inelastic scattering time τ_i in our alloys is estimated from Eqs. (2) and (3), the relation $\Delta\sigma=e^2(D\tau_i)^{-1/2}/(2\pi^2\hbar)$, and the values of D found from the low-temperature range (Table I). Table II lists all the values of τ_i which are $(2.1\text{–}14.3)\times\ 10^{-10}\ T^{-2}$ (s) for $T<\Theta_D/3$ and $(7.7\text{–}20.5)\times\ 10^{-13}T^{-1}$ (s) for $\mathrm{T}>\Theta_D/3$.

Many experimental investigations[8,14] found τ_i of the order of $2.0\times10^{-10}\ \mathrm{T}^{-2}$ (s) in Cu-Zr, Cu-Hf, and Cu-Ti systems in the low-temperature range and they agree quite well with our estimated values. Generally the elastic scattering time (τ_0) is of the order $\sim10^{-15}-10^{-16}$ s in these highly resistive alloys. This is much smaller than the inelastic scattering time ($\tau_i\sim10^{-12}\text{–}10^{-14}$ s) even in the higher temperature range. This strongly suggests a finite probability of electron localization in our alloys.

We thank Professor D. G. Naugle of Texas A&M University for providing the samples and the Department of Science and Technology, Government of India, for financial assistance through Project No. SP/S2/M-24/93.

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