Electron-phonon dephasing time due to the quasistatic scattering potential in metallic glass CuZrAl

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We have measured the electron-phonon scattering time in the metallic glass CuZrAl between 0.37 and 16 K, using the weak-localization magnetoresistance. The electron dephasing rate reveals a T^2 temperature dependence for almost two decades of temperature and is in good quantitative agreement with the theory considering the interference of electron-phonon and elastic electron scattering mechanisms in the presence of quasielastic scattering potential. In addition, this result demonstrates that the Pippard ineffectiveness condition for electron-phonon scattering is not applicable to amorphous alloys that comprise atoms with considerably different atomic masses.

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Recently, intense attention has been paid to the newly found metallic glass CuZrAl in the study of metallic-glass forming ability,[1] specific heat,[2] and resistivity.[3] Since amorphous alloys are typical disordered systems, it would be of interest to investigate the quantum-interference (e.g., weak-localization) transport in CuZrAl. Previous magnetotransport experiments found that the electron dephasing rate in the parent metallic glasses [$Cu_{60}Zr_{40}$ (Ref. 4) and $Cu_{57}Zr_{43}$ (Ref. 5)] revealed an approximate T^2 temperature dependence, but the results had not been fully explained, partly due to the unavailability of an adequate electron-phonon (*e*-ph) interaction theory for disordered conductors at that time. Theoretical studies of the *e*-ph interactions in *disordered* conductors (where $ql<1$, with q being the wave number of thermal phonons, and l being the electron elastic mean free path) and their applicability to experiments have long been a subject of discussion.[6–9] Recently, Sergeev and Mitin[10] have proposed that the *e*-ph interactions are *drastically modified* by the presence of inelastic scattering from quasistatic electron scatterers and compared their calculations with the conventional results of Pippard,[6] Schmid,[7] and Rammer and Schmid.[8] (In those conventional theories,[6–8] the impurities are taken to always vibrate in phase with the deformed lattice atoms.) Essentially, for a disordered conductor, the conventional theory predicts a *weakened* *e*-ph scattering rate given by $\tau_{ep}^{-1}\propto(\tau_{ep}^{0})^{-1}(ql)\propto T^4 l$, while the Sergeev-Mitin theory predicts an *enhanced* scattering rate $\tau_{ep}^{-1}\propto(\tau_{ep}^{0})^{-1}(ql)^{-1}\propto T^2 l^{-1}$, where $(\tau_{ep}^{0})^{-1}\propto T^3$ is the *e*-ph scattering rate in the pure case. Therefore, $(\tau_{ep}^{-1})_{\text{Pippard}}/(\tau_{ep}^{-1})_{\text{Sergeev}}\approx(ql)^2\approx 10^{-4}$ for $ql\approx 0.01$ (which is pertinent to our case), strongly indicating that interference of *e*-ph and elastic electron scattering mechanisms in the presence of quasielastic scattering potential, as proposed by Sergeev and Mitin, can dramatically change the nature of the *e*-ph interaction. Taking $\tau_{ep}^{0}\approx 100$ ns for pure Cu at 1 K,[11] one obtains $(\tau_{ep})_{\text{Sergeev}}\approx 1$ ns for $ql\approx 0.01$, which is in good quantitative agreement with what we measured in the metallic glass CuZrAl (see below). In this case, the atomic masses of Cu (63.5), Zr (91.2), and Al (27) are *distinctly different*, and hence quasistatic electron scattering should be significant, fully justifying the applicability of the Sergeev-Mitin theory.

Ingots of four metallic glasses *A*1-2 ($Cu_{60.3}Zr_{37.2}Al_{2.5}$), *A*2-2 ($Cu_{59.6}Zr_{36.9}Al_{3.5}$), *A*3-2 ($Cu_{58.7}Zr_{36.3}Al_5$), and *A*4-2 ($Cu_{58.1}Zr_{35.9}Al_6$) were obtained by arc melting of a mixture of high purity Cu (99.99 wt. %), Zr (99.9 wt. %), and Al (99.99 wt. %). Bulk metallic-glass rods with a diameter of 3 mm were then prepared by means of copper mould suction casting. The glassy phase of the prepared samples was assessed by examining their x-ray spectra recorded using a rotating-anode x-ray generator (Cu $k\alpha$, 50 kV, 120 mA) with a graphite (002) monochromator. The XRD patterns of the powdered samples all exhibited a single peak and confirmed that all the samples were in a glassy state, Fig. 1.

For resistance and magnetoresistance measurements, the rods were cut into small slices and sanded. The slices have widths of about 1 mm and thicknesses of around 0.5 mm. Electrical contacts of fine Pt wires were glued by silver paint and then baked at 100 °C on a hot plate for 1 h. Magnetoresistances were measured in an Oxford Heliox ^{3}He fridge equipped with a superconducting magnet. Resistances from

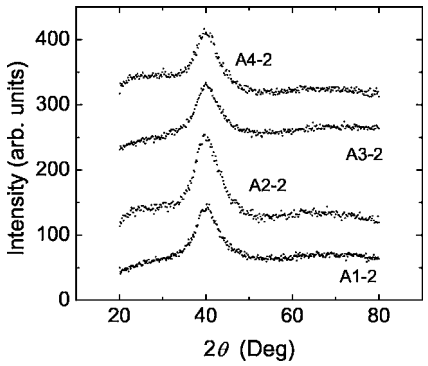

FIG. 1. XRD data for four CuZrAl metallic glasses: *A*1-2, *A*2-2, *A*3-2, and *A*4-2, as indicated.

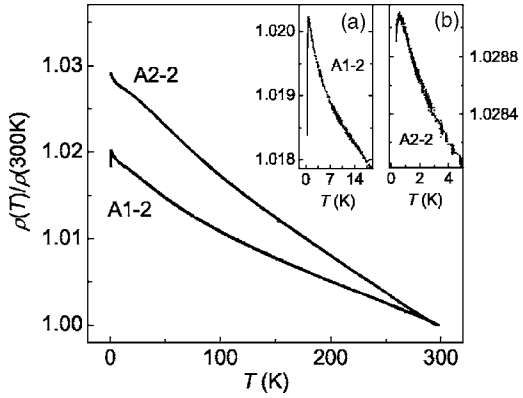

FIG. 2. Normalized resistivity as a function of temperature for samples $A1$-2 and $A2$-2, as indicated. Insets (a) and (b) show the low temperature data in expanded scales. Note that in inset (b) the axis label is on the right-hand side.

0.37 up to 300 K were measured in the same ^{3}He fridge and a standard ^{4}He insert. Four-probe measurements were made by using a Linear Research LR400 resistance bridge operating at 16 Hz and at low enough excitation currents to avoid Joule heating. In this paper the results of two representative samples, $A1$-2 and $A2$-2, are presented.

In zero magnetic field, the resistance of the samples increases as the temperature is lowered from 300 to 1 K. Below 1 K, superconducting onsets were observed in both samples. Figure 2 shows a plot of the variations of the resistance with temperature for the two samples. The onsets of the superconducting temperature, T_c, are 0.83 and 0.76 K for $A1$-2 and $A2$-2, respectively. From 300 to 4 K, the increases in the resistivity are about 2.1% ($A1$-2) and 2.8% ($A2$-2). From 220 to 40 K, and from 9 to 1 K, the resistivities show a $\sqrt{T}$ dependence. From about 30 to 15 K the temperature dependence is essentially linear. The overall temperature behavior of the resistance is similar to that found in $(Cu_{59.6}Zr_{36.9})_{1-x}Al_x$, with $x=0-0.20$.[3] These features are typical and in line with the electrical-transport behavior of amorphous metals when electron-electron interaction and/or quantum-interference effects are the dominant contributions to resistivities.[12,13]

To determine the electron diffusion constant, D, the specific heat was measured on sample $A2-2$ at low temperatures and the electronic contribution γT was obtained, with $\gamma=2.83$ mJ/mol K^2. From the expression $\gamma T=\pi^2 k_B^2 N(0)T/3$ and the Einstein relation $1/\rho=De^2N(0)$, where $N(0)$ is the electronic density of states at the Fermi level, D may be evaluated. According to Li *et al.*,[2] the temperature dependence of specific heat is almost the same for the CuZrAl metallic glasses with various Al atomic contents from 4% to 10%. In our samples the difference of Al content is only 1 at. %, the atomic contents of Cu and Zr differ by even less than 1%. Thus, it is justified to assume that these two samples have basically identical values of γ and therefore $N(0)$. We have also used the measured $\rho(4\text{ K})$ to compute the values of D and found similar results. The values of D together with values of relevant parameters for our samples are listed in Table I.

The values of k_Fl (k_F is the Fermi wave number) may be determined from the expression $k_Fl=3mD/\hbar$ (m is the free-electron mass) and are listed in Table I. Taking the Fermi energy $E_F=1.3$ eV for the $Cu_{50}Zr_{50}$ amorphous alloy,[14] and using the average value of $k_Fl\approx1.13$, we obtain $l\approx1.9$ Å in our samples. This size of l is common in amorphous alloys and is comparable with the atomic spacing of the materials.

TABLE I. Values of relevant parameters for samples $A1$-2 and $A2$-2. The values of diffusion constant D were determined from low-temperature specific heat measurements. ρ is in $\mu\Omega$ cm.

	ρ(300 K)	ρ(4 K)	D (cm^2/s)	k_Fl	τ_{so}^{-1} (10^{11} s^{-1})
$A1$-2	190	194	0.46	1.19	2.6
$A2$-2	211	214	0.41	1.06	1.6

The magnetoresistances of the samples are positive between 0.37 and 16 K (not shown). In order to apply the weak-localization theoretical predictions, we need to examine the value of k_Fl. Strictly speaking, the weak-localization theory is formulated for the condition $k_Fl\gg1$. However, Howson and Gallagher[13] have shown that, in amorphous alloys, the criterion can be relaxed to be $k_Fl>1$. We have analyzed the magnetoresistance data with the three-dimensional weak-localization theoretical predictions, taking into account strong spin-orbit scattering and superconducting fluctuation contributions.[15,16] The details of the least-squares fitting procedure has been discussed previously.[17] The superconducting fluctuation contribution to the magnetoresistance corresponding to the Maki-Thompson term is represented by an adjusting parameter called the Larkin's electron-electron attraction strength β.[16] Our fitted value of β for sample $A1$-2 is comparatively higher than that for sample $A2$-2. This is because the T_c is higher in $A1$-2 than in $A2$-2. From Larkin's expression[16] of $\beta=\beta(T/T_c)$ (the solid curve in Fig. 3), we determine the T_c of $A1$-2 to be 0.20 ± 0.02 K. For sample $A2$-2, it is difficult to determine the magnitude of T_c from the fitted β values. However, we may compare the onset of the superconducting transition for the two samples and estimate that the T_c of $A2$-2 should be around 0.13 ± 0.02 K, which is well below our lowest temperature of measurement.

The electron dephasing time can be calculated from the fitting parameter $B_\varphi=\hbar/4eD\tau_\varphi$ defined in the weak-

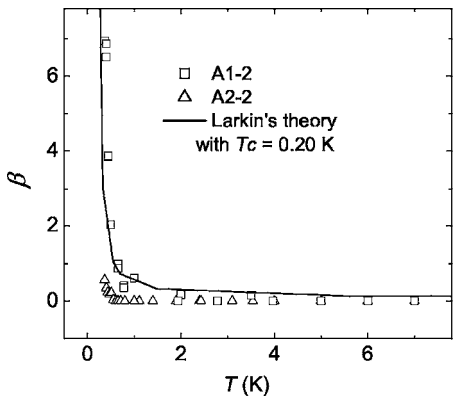

FIG. 3. Electron-phonon coupling strength $\beta(T/T_c)$ as a function of temperature for samples $A1$-2 and $A2$-2. The solid curve is the Larkin's prediction (Ref. 16) of β with a $T_c=0.20$ K.

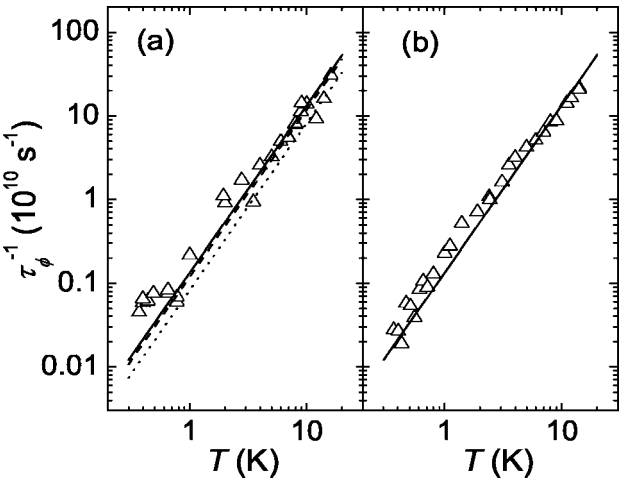

FIG. 4. Dephasing rate as a function of temperature for samples *A*1-2 (a) and *A*2-2 (b). The straight lines are the theoretical predictions of Eq. (1) with the parameter $k=0.3$ (dashed line), 0.48 (solid line) and 0.8 (dotted line) (see text). Note that in (a) and (b) the τ_φ^{-1} axis has identical scales.

localization theory.[12,15] The results of our measured τ_φ^{-1} are presented in Fig. 4 in double logarithmic scales. Close inspection shows that, below about 1 K, the dephasing rate in the sample *A*1-2 [Fig. 4(a)] reveals a somewhat "leveling-off" behavior, reflecting stronger superconducting fluctuation contributions due to a higher T_c in this sample, as compared with that in the sample *A*2-2 where τ_φ^{-1} decreases monotonically with decreasing temperature all the way down to 0.37 K. Analogous leveling-off behavior in τ_φ^{-1} has been found in superconducting materials at temperatures just above T_c.[17–19] Most notably, we see that in the whole temperature range the dephasing rate reveals a linear dependence on T in the double logarithmic scales, with a slope of 2, as indicated by the straight lines in Figs. 4(a) and 4(b). That is, $\tau_\varphi^{-1} \propto T^2$. Such a quadratic temperature dependence of the inelastic dephasing rate can be ascribed to the *e*-ph scattering in the presence of disorder. The *e*-ph scattering time in disordered metals containing both "vibrating" and "quasistatic" impurities has recently been calculated by Sergeev and Mitin,[10] and is given by

$$\frac{1}{\tau_{ep}} = \frac{\pi^4 (k_B T)^4 l}{5\hbar^2 (\hbar k_F)^2} \frac{\beta_l}{v_l^3}\left(1 + \frac{3k}{2}\frac{v_l^5}{v_t^5}\right) + \frac{3\pi^2 (k_B T)^2}{2(\hbar k_F)^2 l}\frac{\beta_l}{v_l}(1-k)\left(1 + 2k\frac{v_l^3}{v_t^3}\right), \quad (1)$$

where $\beta_l = (\frac{2}{3}E_F)^2 N(0)/2\rho_m v_l^2$, and ρ_m is the mass density. $v_l = 4660$ m/s and $v_t = 2120$ m/s are, respectively, the longitudinal and transverse sound velocities.[2] $k \equiv 1 - l/\mathcal{L}$ (where $\mathcal{L}$ is the electron mean free path with respect to the quasistatic potential) is a parameter representing the relative part of the vibrating potential to the total potential. From specific heat measurement, we obtain $N(0) = 1.2$ states/(eV atom). This value is comparable with the value $N(0) = 1.6$ states/(eV atom) calculated for the $Cu_{50}Zr_{50}$ metallic glass.[14] In our experiment, $l = 1.9$ Å and $\beta_l = 0.028$. The theoretical prediction with these material parameters and a value of $k = 0.48$ gives $\tau_{ep}^{-1} = (3.4 \times 10^5 T^4 + 1.34 \times 10^9 T^2)$ s^{-1}, where T is in Kelvin. This prediction is presented by the straight solid lines in Figs. 4(a) and 4(b).[20] A good *quantitative agreement* between our experiment and the theoretical predictions is clearly seen. Therefore, it is obvious that the T^2 term in Eq. (1) is in dominance in the metallic glass CuZrAl, implying the total τ_{ep}^{-1} is essentially determined by the coupling of electrons with transverse phonons while the coupling of electrons with longitudinal phonons is negligibly weak.[10] Sergeev and Mitin have pointed out that for the *e*-ph scattering in the intermediate-disorder regime of $ql \gtrsim 1$, a seemingly T^2 dependence of τ_{ep}^{-1} may be observed.[9,10,21] In our case, the value $q_t l \approx k_B T l/\hbar v_t \approx 0.012T$ (q_t is the wave number of transverse thermal phonons), i.e., $q_t l \ll 1$ and the *e*-ph scattering is in the dirty limit. It should be noted that our measured T^2 dependence is robust and persists for almost two decades of temperature. A T^2 dependence of τ_{ep}^{-1} in the dirty limit has recently been found in *polycrystalline* TiAl,[22] TiAlSn,[23] AuPd,[24] and VAl (Ref. 25) alloys, and Au-doped In_2O_{3-x} films[26] over a somewhat limited temperature range, where no satisfactory quantitative comparison between the theory and experiment was achieved. In the case of the In_2O_{3-x}:Au films, the added Au atoms played the role of the quasistatic electron scatterers. On the other hand, a T^{-4} behavior manifesting the τ_{ep} subject to the Pippard ineffectiveness condition (i.e., long wavelength phonons are ineffective in scattering short mean-free-path electrons[6]) has very recently been observed in *elemental* Cu,[27] Au,[27] Hf,[28] and Ti (Ref. 28) thin films at very low temperatures below hundreds mK.

There are two more free parameters in the weak-localization theory: the spin-orbit scattering time, τ_{so}, and the spin-spin scattering time, τ_s. In this study, we found that τ_s^{-1} is negligibly small in our samples. This is in consistence with the theoretical understanding that the Maki-Thompson superconducting fluctuation contribution would be suppressed in the presence of spin-spin interaction. The values of τ_{so}^{-1} in our samples are on the order of 10^{11} s^{-1} (see Table I). At the high temperatures of our measurement, the values of τ_{so}^{-1} are comparable with τ_{ep}^{-1}. At the low temperatures of our measurement, they are 2 to 3 orders of magnitude larger than τ_{ep}^{-1}. This is the reason why the magnetoresistances are always positive in our case. (The Maki-Thompson superconducting fluctuation term also contributes a positive magnetoresistance.)

We should like to compare our data with previous works on similar materials. In the film metallic glasses $Cu_{60}Zr_{40}$ studied by Poon *et al.*[4] and $Cu_{57}Zr_{43}$ studied by Bieri *et al.*,[5] the temperature dependent inelastic electron scattering rates were extracted from weak-localization magnetoresistance. Poon *et al.* obtained an inelastic rate $\tau_{in}^{-1} \approx 10^9 T^{2.2}$ s^{-1}, while Bieri *et al.* reported an τ_{in}^{-1} varied approximately as T^2 above 4 K. Since the electrical-transport parameters in these two experiments are comparable with the parameters in our bulk metallic glasses, we suspect that the inelastic rate τ_{in}^{-1} found in Refs. 4 and 5 may be safely ascribed to the *e*-ph scattering rate and satisfactorily explained by Eq. (1).

In conclusion, we have measured the magnetoresistance

of the newly found metallic glass CuZrAl and extracted the electron dephasing time. This work demonstrates that the Pippard ineffectiveness condition for *e*-ph scattering is not applicable to amorphous alloys that comprise atoms with *considerably different* atomic masses. Our result shows the fast *e*-ph dephasing rate with T^2 temperature dependence, which corresponds to the interference of *e*-ph and elastic electron scattering mechanisms in the presence of quasistatic scattering potential. Moreover, our results are in good *quantitative agreement* with the Sergeev-Mitin interference theory of the *e*-ph interaction in disordered conductors.

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