Measurement of low-temperature transport properties of Cu-based Cu-Zr-Ti bulk metallic glass

Y. K. Kuo* and K. M. Sivakumar
Department of Physics, National Dong Hwa University, Hualien 97401, Taiwan

C. A. Su, C. N. Ku, and S. T. Lin
Department of Physics, National Cheng Kung University, Tainan 70101, Taiwan

A. B. Kaiser
MacDiarmid Institute for Advanced Materials and Nanotechnology, Victoria University of Wellington, P.O. Box 600, Wellington, New Zealand

J. B. Qiang, Q. Wang, and C. Dong
State Key Laboratory of Materials Modification & Department of Materials Engineering, Dalian University of Technology, Dalian 116024, China

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The transport properties, including electrical resistivity (ρ), thermopower (S), and thermal conductivity (κ) of bulk metallic glass alloys $Cu_{64}Zr_{28.5}Ti_{7.1}$, $Cu_{62.3}Zr_{23.7}Ti_{10}$, $Cu_{60.6}Zr_{26.9}Ti_{12.5}$ and $Cu_{58.8}Zr_{26.2}Ti_{15}$ in the temperature range 10–300 K are reported. The temperature variations of electrical resistivity in these alloys, with a negative temperature coefficient, are found to be rather weak. These findings are consistent with the metallic glass nature of these compounds. It is observed that the electrical resistivity increases with increasing Ti concentration, ascribed to the enhancement of disorder with such a composition change. The magnetoresistivity of $Cu_{64}Zr_{28.5}Ti_{7.5}$ alloy decreases with increasing temperature and increases with increasing magnetic field, suggesting that the weak localization effect dominates the electrical transport. The temperature-dependent thermopower and thermal conductivity characteristics are nearly identical and weakly independent of compositions. It is noted that the observed $\kappa(T)$ of Cu-Zr-Ti metallic glass alloys show notable similarities with the quasicrystalline system. There are two main features in $S(T)$, a knee around 120 K and a plateau below 50 K, which represent the deviation from the expected linear behavior. A detailed theoretical analysis has suggested that the appearance of the knee is due to the electron-phonon interaction at low temperatures and the plateau is associated with low-energy excitations in glasses.

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I. INTRODUCTION

In the past two decades, bulk metallic glass (BMG) alloys have occupied an important position due to their unique physical properties[1–9] and technological importance.[10–12] With minimum crystal defects, BMGs are the imminent candidates to replace the conventional materials (like steel, ceramic, etc.) used in the industry as a result of their superior mechanical properties, for example, strength, hardness, toughness, and elasticity. Further, low eddy current loss with soft magnetic properties of magnetic BMGs turns out to be an additional advantage of these technologically key classes of alloys. Metallic glass possesses a glassy structure with local atomic configurations, a high degree of dense random packing, and long-range homogeneity, which are very different from those of the corresponding crystalline phases. Therefore, despite the significant volume of investigations, the transport properties of BMG are still controversial.

In general, the BMG alloys can be classified into two types, i.e., magnetic and nonmagnetic. Particularly, the nonmagnetic bulk metallic glass (NBMG) alloys are of significant experimental and theoretical interests due to their peculiar low-temperature transport properties.[4] These intriguing physical properties are different from those of crystalline solids but show striking similarities with amorphous materials. Especially below room temperature, the temperature-dependent characteristics of these transport properties were found to be universal to the glassy structure rather than their chemical composition.[13–17] Regarding the electrical transport, a large value of magnetoresistance and a positive but small value of Hall coefficient with significant temperature dependence have been observed in some of the nonmagnetic BMG alloys.[9] These observations indicate the inadequacy of the earlier proposed nearly free electron model, which is usually employed while explaining the transport properties of amorphous metals.[9] In addition, the residual resistivity of the NBMG is considerably larger than that of the crystalline counterpart.[1] Another remarkable feature of the NBMG, known as the Mooij criterion, is the correlation between the temperature coefficient of resistivity (TCR) and residual resistivity $\rho(0)$. Usually, the TCR of BMG alloys with $\rho(0) > 150\ \mu\Omega$ cm is negative and turns to positive as $\rho(0)$ falls below 150 $\mu\Omega$ cm. A systematic annealing study above and below the crystallization temperature could result in a gradual sign reversal in TCR.[1]

Thermopower (S) is another transport property, which also shows distinct behaviors for BMG.[2,5,6] The near-linear characteristics reported in $S(T)$ around room temperature are usually unique to a glassy/amorphous system. Such a behavior is generally absent in the crystalline materials, although the simple theories predict linear temperature dependence.[9]

At temperatures below the Debye temperature, the deviation from the linear behavior has been attributed to the electron-phonon mass enhancement.[9,18] In general, BMG alloys with low electrical resistivity (less than 150 $\mu\Omega$ cm) and positive TCR show negative thermopower, while the alloys with higher resistivity (>150 $\mu\Omega$ cm) and negative TCR show positive thermopower.[2,9] Moreover, a clear deviation from the Debye's theory, which is expected to follow T^3 dependence, was observed in the low-temperature specific heat of BMG.[4,16] This deviation is attributed to the difference or excess in the vibrational density of states over the crystalline phase, namely, the boson peak. A comparative approach in interpreting the resistivity and thermopower between Ziman and Mott models has revealed that neither model gives correct resistivities, but the Mott model provides a reasonable description to the thermopower of BMG alloys.[19] For some of the amorphous metals, the sign and magnitude of thermopower are found to be a strong function of composition. To explain this behavior, an extended Ziman model (Faber-Ziman) has been effectively introduced to take account of the different way in which the various constituents contribute to the measured thermopower.[20,21] In the case of NBMG alloys, the larger phonon wavelength and rapidly dropping number of phonons lead to very ineffective electron-phonon scattering at low temperatures,[19] and hence phonon drag contribution becomes insignificant. Nevertheless, the appearance of a "knee" around 50 K in temperature-dependent thermopower has been reported in many of the NBMG alloys,[19,20] which has been also correlated to the electron-phonon mass enhancement. The reported electron-phonon mass enhancement was found to decrease as the temperature increases and completely disappears at temperatures comparable with the Debye temperature. Inclusion of such an additional effect, electron-phonon mass enhancement, resulted in a good description of the thermopower of amorphous transition metal alloys.[19]

Recently, a Cu-based $Cu_{60}Zr_{30}Ti_{10}$ BMG alloy was found to have good mechanical properties in addition to their extremely high glass-forming abilities and expected to be developed as a new type of structural material with higher strength combined with good ductility.[12,22] A previous report on the similar class of NBMG alloy Cu-Zr-Al has indicated the presence of low-temperature anomalies in the specific heat.[4] In the present work, we have carried out electrical resistivity (ρ), thermopower (S), thermal conductivity (κ), and magnetoresistivity (MR) measurements on nonmagnetic BMG alloys Cu-Zr-Ti from 10–300 K. These alloys have been shown to have high glass-forming abilities.[23] The purpose of the present study is to probe the changes in the scattering mechanism of the charge carriers with respect to composition change through these transport properties. The thermopower measurement on NBMG alloys is much easier to interpret than the magnetic BMG and crystalline alloys due to the absence of contributions from magnetic excitations and insignificant phonon drag effect.[5,19] To the best of our knowledge, these measurements represent one of the most comprehensive transport studies on this class of BMG alloys at low temperatures.

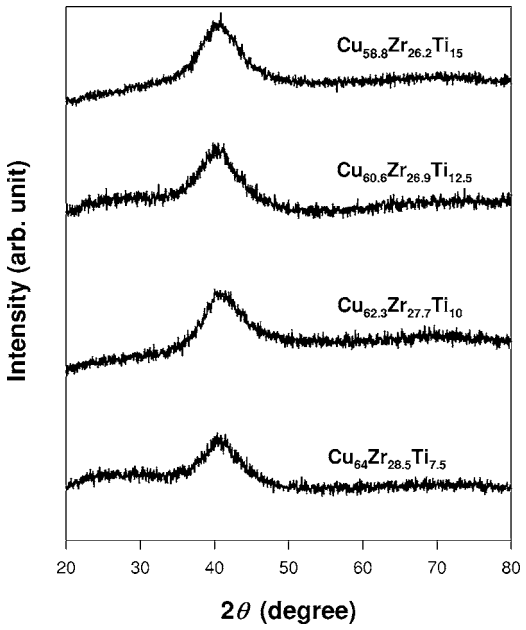

FIG. 1. The x-ray-diffraction patterns of Cu-Zr-Ti alloys. The glassy nature in these alloys is clearly seen.

II. EXPERIMENT

Ingots of the $(Cu_{9/13}Zr_{4/13})_{100-x}$-$Ti_x$ serial BMGs $Cu_{64}Zr_{28.5}Ti_{7.5}$, $Cu_{62.3}Zr_{23.7}Ti_{10}$, $Cu_{60.6}Zr_{26.9}Ti_{12.5}$, and $Cu_{58.8}Zr_{26.2}Ti_{15}$ were obtained by arc-melting of a mixture of high-purity Cu (99.99 wt %), Zr (99.9 wt %), and Ti (99.99 wt %). Bulk metallic-glass rods with a diameter of 3 mm were then prepared by means of copper mold suction casting. Figure 1 shows the x-ray diffraction (XRD) patterns of the prepared samples. All the XRD patterns exhibit a single broad peak, indicating the glassy phase of the samples. Electrical resistivity measurements were performed by a standard four-point probe method from 10 to 300 K and the magnetoresistivity was measured under a magnetic field H from 0 to 6 T at different temperatures. The thermal conductivity and thermopower measurements were simultaneously performed by a heat pulse technique in a helium closed-cycle refrigerator. It is well known that the values of thermopower for BMG alloys are very small. Therefore, we have carefully calibrated the leads with a YBCO superconductor below the superconducting transition temperature (~90 K) for reliable and accurate measurement at low temperatures. Details of the experimental technique can be found elsewhere.[24]

III. RESULTS AND DISCUSSION

A. Electrical resistivity and magnetoresistivity

The temperature-dependent electrical resistivity $\rho(T)$ of the BMG alloys $Cu_{58.8}Zr_{26.2}Ti_{15}$, $Cu_{60.6}Zr_{26.9}Ti_{12.5}$, $Cu_{62.3}Zr_{27.7}Ti_{10}$, and $Cu_{64}Zr_{28.5}Ti_{7.5}$ is shown in Fig. 2. The room temperature ρ values of these measured alloys were found to be between 190 and 270 $\mu\Omega$ cm. A weak temperature variation and a negative temperature coefficient in resistivity (TCR) were observed. These features are in accordance with the Mooij criterion for NBMG alloys.[9] It was noticed

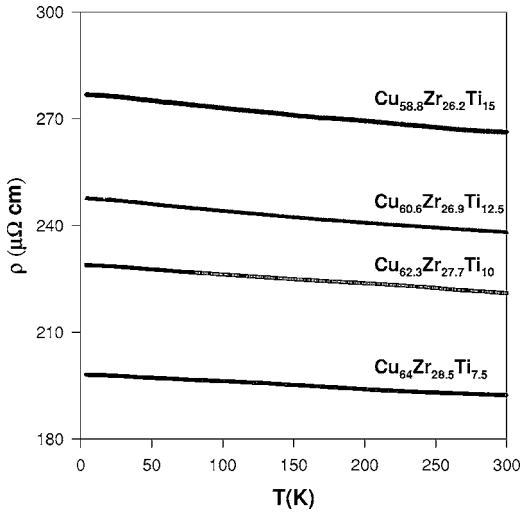

FIG. 2. Electrical resistivity as a function of temperature of Cu-Zr-Ti alloys with various compositions.

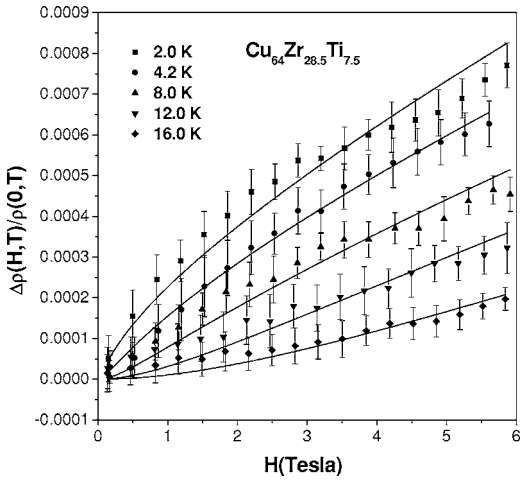

FIG. 3. The MR, $\Delta\rho(H,T)/\rho(0,T)$, as a function of magnetic field H at different temperatures for $Cu_{64}Zr_{28.5}Ti_{7.5}$. Solid curves are the theoretical fits.

that an increase in Ti concentration (or a decrease in Cu/Zr concentration) leads to a systematic increase in ρ, most likely due to the enhanced disorder with increasing Ti concentration. The addition of the third element Ti to the Cu-Zr system enhances the disorder and hence results in an increase in the electrical resistivity. This is further evident from the corresponding increase in the residual resistivity $\rho(0)$ for higher Ti concentrations. In such a scenario, the increase in ρ would saturate when the disorder reaches its maximum level for a given concentration of Cu, Zr, and Ti. Earlier reports on similar classes of alloys (Cu-Zr and Cu-Ti) with varying concentrations of Cu/Ti/Zr have revealed only a slight variation in ρ, and a saturation in residual resistivity was indeed observed (see Ref. 9, and references therein).

Many scattering mechanisms have been suggested to explain the observed electrical transport, particularly the negative temperature coefficient found in the electrical resistivity.[1,2,5,25,26] For example, the mechanism proposed by Ziman, originally meant for liquid transition metals, takes into account the scattering from the structural disorder,[25] while the Mott *s-d* scattering theory considers the thermal broadening of the electron distribution and the shifting of the chemical potential in explaining the sign of TCR.[26] Another mechanism for the negative sign of the temperature dependence of resistivity in highly disordered materials (typically those with resistivities above 150 $\mu\Omega$ cm [Mooij criterion]) is weak localization. In this scenario, the increased resistivity due to the coherence of elastically backscattered carriers is destroyed by inelastic scattering by phonons as the temperature increases, leading to a progressive reduction in resistivity.[27,28] Figure 3 shows the magnetoresistivity (MR), $\Delta\rho(H,T)/\rho(0,T)$, as a function of magnetic field H at different temperatures for $Cu_{64}Zr_{28.5}Ti_{7.5}$. It is seen that the MR of the $Cu_{64}Zr_{28.5}Ti_{7.5}$ alloy decreases with increasing temperature and increases with increasing magnetic field. Such a behavior follows the description of weak localization effect including strong spin-orbit interaction for disordered systems.[29] As seen in Fig. 3, the MR data can be fitted fairly well with an inelastic scattering time $\tau_i = \tau_0$, T^p with $\tau_0 \approx 5\times10^{-9}$ s, $p \approx 2.5$, and spin-orbit scattering time $\tau_{so} \approx 1\times10^{-13}$ s, by using the theories of electron-electron interaction[30] and weak localization[29] in a magnetic field. The value of $p \approx 2.5$ suggests the inelastic electron scattering process at low temperatures is dominated by electron-phonon scattering.[31,32] The details of our fitting procedure can be found elsewhere.[33]

B. Thermopower

The temperature-dependent thermopower $S(T)$ of Cu-Zr-Ti BMG alloys is shown in Fig. 4. The room temperature S values for studied alloys were found to be small, around 2–3 μV/K. Such a small value of S is generally observed in NBMG alloys. It is noted that S remains positive in the entire temperature range of investigation, in accordance with the Mooij-type correlation between the magnitude of electrical resistivity and the sign of thermopower.[2,9] The observed

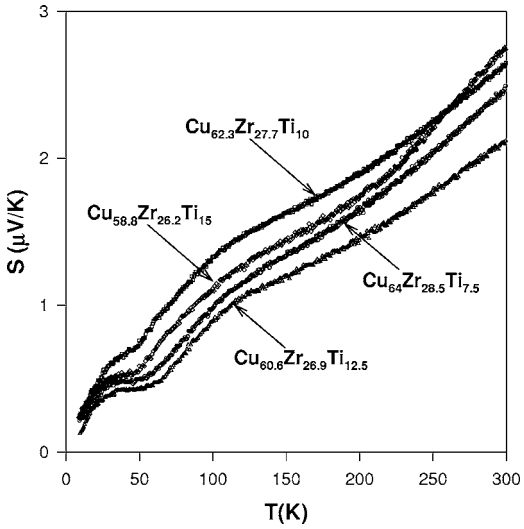

FIG. 4. Temperature dependence of the thermopower of Cu-Zr-Ti alloys with various compositions.

magnitude and temperature-dependent characteristics in S are nearly identical for all the compositions presently investigated. In fact, the change in Cu/Ti concentration is expected to have a corresponding effect on the electronic structure and hence changes in S, as it is well known that S is very sensitive to the changes in the DOS near the Fermi level.[2,5,18] However, no apparent composition-dependent change in S with respect to the Cu/Ti concentration could be noticed, suggesting that the changes in the electronic structure near the Fermi level are masked from the influences owing to the changes in the composition.

This is similar to the case of Be-based ternary alloys, in which the resonant levels Cu, Ti, and Zr are well above (below) the Fermi level[7] and hence neither the shifting of Fermi level nor the altering of DOS near the Fermi level takes place. Other consequences, such as changes in the local atomic environment around Cu/Ti atoms may also play a dominant role in the electronic structure, but still show no significant effect on their bulk thermoelectric transport property. Unlike the electrical resistivity, increased disorder shows no significant effect on thermopower in these alloys. According to the Mooij-type relationship, the sign of S changes from positive to negative when the value of $\rho(0)$ falls below 150 $\mu\Omega$ cm. In our present study, the values of $\rho(0)$ for all studied Cu-Zr-Ti BMG alloys are higher than 200 $\mu\Omega$ cm and the values of S are indeed positive, in agreement with the Mooij criterion. Another noticeable feature is that the observed S is almost independent of composition in these alloys. This finding implies that beyond a certain degree of disorder, S may remain unaffected or reach saturation. Such an intriguing correlation is actually observed in the wide variety of NBMG alloys, suggesting that the characteristics of temperature-dependent thermopower are governed by the disorder rather than their individual electronic structure or chemical composition.[9]

The temperature dependence of the thermopower of these Cu-Zr-Ti BMG alloys also exhibits intriguing features. The observed trend of increasing thermopower as temperature increases is clearly indicative of metallic diffusion thermopower, while the existence of a knee at around 100 K with an increased gradient at lower temperatures suggests the effect of electron-phonon enhancement of diffusion thermopower at low temperatures.[18,34,35] This effect is typically seen in highly disordered metallic materials with resistivities greater than 100 $\mu\Omega$ cm where the phonon drag thermopower is strongly suppressed,[35] so the measured resistivities of around 200 $\mu\Omega$ cm or more for our BMG alloys (Fig. 2) suggest that the electron-phonon enhancement effect in thermopower would be expected to be observable in these materials.

We have therefore compared our thermopower data to the theoretical expression for the temperature-dependent metallic diffusion thermopower S_d including the electron-phonon enhancement effect;[18]

$$S_d = S_u[1 + a\bar{\lambda}(T)], \tag{1a}$$

$$\bar{\lambda}(T) = \frac{2}{(k_B T_D^*)^2}\int_0^{k_B T_D^*} E G_S(E/k_B T)dE. \tag{1b}$$

Here S_u is the unenhanced diffusion thermopower, usually taken as linear in temperature ($S_u = bT$) which is the usual

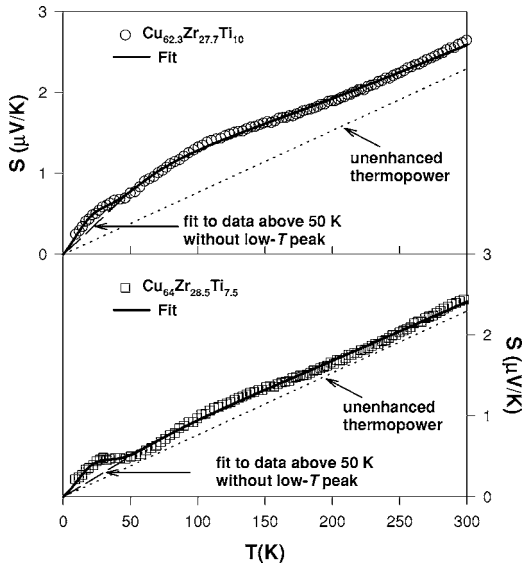

FIG. 5. Fits of Eq. (2) including electron-phonon enhancement and a low-temperature peak (solid lines) to the measured thermopower of the Cu-Zr-Ti BMG alloys with 7.5% and 10% Ti; also shown are the unenhanced diffusion thermopowers (thin dotted lines) for the fits, and fits to the data above 50 K to Eq. (1) omitting the low-temperature peak (dash-dot lines). Parameter values are discussed in the text.

Mott approximation for metallic diffusion thermopower.[9,33] The second factor, within brackets in Eq. (1a), describes the enhancement of the thermopower at low temperatures by the electron-phonon interaction, which is analogous to the well-known electron-phonon enhancement of electronic specific heat at low temperatures. The parameter a gives the magnitude of the enhancement factor for diffusion thermopower in the low-temperature limit. The temperature dependence of this enhancement is given by $\bar{\lambda}(T)$, given by Eq. (1b), where k_B is Boltzmann's constant and $G_S(E/k_BT)$ is the universal thermopower enhancement function we have defined earlier.[18,32] Note that $\bar{\lambda}(T)$ is normalized so that $\bar{\lambda}(0)=1$. This simple form of the expression for $\bar{\lambda}(T)$ assumes a Debye-type Eliashberg function to describe the interaction of electrons and phonons, with $k_BT_D^*$ an effective Debye cutoff energy for the electron-phonon enhancement effect, but the shape of the calculated thermopower is rather insensitive to the precise shape of the phonon spectrum assumed.[18] The decrease of the enhancement $\bar{\lambda}(T)$ at higher temperatures gives rise to a characteristic knee or change of slope in S_d which is typically centered around a temperature of approximately $0.25T_D^*$.[35]

Equation (1) does give a reasonable description of the measured thermopower of the Cu-Zr-Ti BMG alloys above 50 K. This is illustrated in Fig. 5 for the 7.5% Ti and 10% Ti data, where the dash-dot line is the fit of Eq. (1) to the data above 50 K (this fit essentially coincides with the full line at temperatures above 50 K). The knee in the data just above a temperature of 100 K, and the fact that the data above 100 K do not extrapolate to zero thermopower at zero temperature, do appear compatible with the electron-phonon enhancement of diffusion thermopower.

However, Eq. (1) cannot account for the additional plateaulike effect that is seen below 50 K in all the Cu-Zr-Ti BMG alloys. The discrepancy between the data below 50 K and the fit to Eq. (1) suggests the presence of an additional low-temperature thermopower peak contribution, which combined with the linear increase of diffusion thermopower at low temperatures yields the observed plateaulike behavior. We model this contribution by adding a Gaussian peak in Eq. (1a) as follows:

$$S = bT\{1 + a\bar{\lambda}(T) + c\exp[-(T-f)^2/(2w^2)]\}, \qquad (2)$$

where the peak is determined by the new parameters c, f, and w (note that adding the Gaussian peak in this way rather than directly in S ensures that S goes to zero at $T=0$ as required for thermopower). The most obvious origin of this thermopower peak contribution is a small remnant phonon drag term—since the thermopower of our Cu-Zr-Ti samples is very small, a phonon drag peak more than an order of magnitude smaller than that in typical crystalline metals[36] would still be easily seen. However, the existence of additional low-energy vibrational states (the boson peak) in glassy materials[12,14] could also play a role.

Two examples of the fits of Eq. (2) to the thermopower data for the Cu-Zr-Ti BMG alloys are shown in Fig. 5. It can be seen that the addition of the low-temperature peak term to the enhanced diffusion thermopower of Eq. (1) leads to a good description (shown by the solid line) of the plateaulike behavior below 50 K. The fit parameters c, f, and w for this peak term are quite similar for the four samples and do not show monotonic variations as a function of Ti concentration, so it is clear we are seeing essentially the same effect for each of the samples. The average value and the standard deviation of these parameters for the four samples are $c=1.23\pm0.07$, $f=14\pm3$ K, and $w=12.7\pm0.5$ K. Taking into account the T factor in Eq. (2), the maximum thermopower contribution of this term is 0.17 ± 0.01 μV/K and occurs at a temperature of 22 ± 3 K. This value for the peak magnitude emphasizes that this is a very small thermopower effect.

Above 50 K, the thermopower peak term makes a negligible contribution to the calculated thermopower fit, which in the range 50–300 K is determined solely by the enhanced diffusion thermopower term given in Eq. (1) with fitting parameters a, b, and T_D^*. It can be seen from Fig. 5 that the linear unenhanced thermopower terms in the fits for the 7.5% and 10% Ti samples are almost identical (with linear coefficient $b=7.7$ nV/K^2). The average value and the standard deviation of the fit parameters for the four samples are $b=7.6\pm0.9$ nV/K^2, $a=0.51\pm0.25$, and $T_D^*=440\pm120$ K. The magnitude of the electron-phonon enhancement was larger for the 10% Ti sample ($a=0.9$) than for the others ($a=0.3$–0.5), but the average value is similar to that observed in thin films of splat-quenched Cu-Zr.[34] The fit values of the effective Debye temperature T_D^* are larger than the values of Debye temperature estimated from measurements of specific heat and other properties of Cu-Zr and Cu-Ti films,[34] suggesting that the electron-phonon interaction giving rise to the thermopower enhancement in our Cu-Zr-Ti BMG alloys could be stronger for higher-energy phonons.

We note in Fig. 5 an upturn in the experimental data above the theoretical curve at room temperature, which is likely due to deviations from linearity in the variation of electronic band parameters near the Fermi energy that affect the unenhanced thermopower as temperature increases and the Fermi window expands away from the Fermi level. This deviation is strongest in the Cu-Zr-Ti BMG alloy with greatest Ti concentration (15%), but otherwise the fit is similar to that for the 7.5% Ti sample.

An additive low-temperature peak term such as that in Eq. (2) is appropriate if the peak is due to phonon drag, since drag and diffusion thermopower contributions to thermopower are additive.[36] An alternative interpretation could be that the low-temperature peak and the knee at higher temperatures could be due to different and multiplicative enhancement effects, which would suggest the form

$$S = bT\{1 + a\bar{\lambda}(T)\}\{1 + c\exp[-(T-f)^2/(2w^2)]\} \qquad (3)$$

for the total thermopower. We have tested this expression by fitting it to the data for the 7.5% Ti sample, obtaining a fit indistinguishable to that shown in Fig. 5.

The parameter values for a, b, and T_D^* agreed with those for the fit to Eq. (2) to within 0.1%; this is because these parameters are determined by the thermopower at the knee near 120 K and higher temperatures where the second enhancement factor in Eq. (3) is equal to 1. Likewise, the parameters f, w, and $(1+a)c$ agreed with the values of f, w, and c for the fit to Eq. (2) to within 1%; this reflects the fact that the peak term is significant only below 50 K where the electron-phonon enhanced thermopower is simply $S=bT(1+a)$ to a good approximation. So, we conclude that the data are equally well described by Eq. (3).

C. Thermal conductivity

The temperature-dependent thermal conductivity $\kappa(T)$ for Cu-Zr-Ti alloys is shown in Fig. 6. The room temperature values of the alloys presently investigated lies between 8 and 10 W/m K, which is comparable with other reported results for NBMG alloys.[3,8,37] Similar to that observed in thermopower, magnitude and characteristics in thermal conductivity are nearly identical and independent of composition. Since the thermal conductivity of bulk metallic glass is expected to follow the Wiedemann-Franz law, the electronic part of thermal conductivity can therefore be estimated by using $\kappa_e\rho/T=L_0$. Here ρ is the dc electrical resistivity and the Lorenz number $L_0=2.45\times10^{-8}$ W Ω K^{-2}. As illustrated in Fig. 6, the dotted lines represent calculated κ_e for each sample. From this estimation it could be seen that the phonon contribution is substantial (nearly equal to electronic contribution) in the presently investigated Cu-Ti-Zr alloys, in contrast to the general impression that the glassy material generally has small lattice thermal conductivity.

As the temperature decreases, κ starts to decrease quasilinearly down to about 30 K, and then develops into a shallow plateau, which is followed by a steep fall (T^2 dependence). Such a temperature variation in thermal conductivity is a typical characteristic of glassy materials.[15,16,38] The plateau and the subsequent T^2 fall in κ at low temperatures have

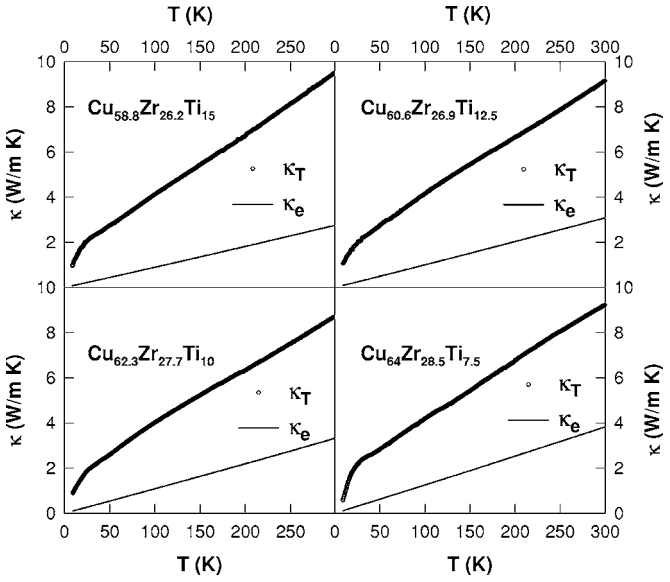

FIG. 6. Temperature dependence of the total thermal conductivity of Cu-Zr-Ti alloys with various compositions. The solid lines represent the calculated electronic contribution.

been attributed to the combined scattering of tunneling and soft vibrational states.[38] Several attempts have been made to correlate the similarities in $\kappa(T)$ between the quasicrystalline and glassy-amorphous alloys.[39–42] The temperature-dependent thermal conductivity for these systems typically shows a monotonic increase with temperature at high T, the appearance of a plateau between 10 and 30 K, and nearly identical variations below the plateau. Further, the temperature-dependent characteristics were found to be almost independent of their chemical composition. Actually a wide variety of amorphous materials, for example, disordered crystals and polymers show similar temperature-dependent characteristics in their thermal conductivity, indicating that the thermal conductivity of amorphous solids is independent of chemical composition and structural details.[13–17] The appearance of a plateau in the thermal conductivity, one of the most prominent features for amorphous solid, is thought to be the consequence of a rapidly decreasing phonon mean-free-path with the increasing frequency.[43]

In fact, the low-temperature plateau in quasicrystals is generally observed at a slightly higher temperature than that of the glassy-amorphous alloys.[38–48] Nevertheless, the thermal conductivity for quasicrystals and amorphous solids-metallic glasses exhibits intriguing similarities above the plateau, i.e., the monotonic increase with temperature.[40,42] The internal friction and thermal conductivity measurements on a single grain of icosahedral Al-Pd-Mn quasicrystal strongly suggest the existence of glasslike low-energy excitations.[42] The common feature between the various amorphous solids is their aperiodicity, while the quasicrystals differ from the amorphous solids by their quasiperiodicity. Michalski *et al.*[39] have performed a theoretical calculation of periodic (crystalline), aperiodic (amorphous), and quasiperiodic (quasicrystalline) models and suggested that the monotonic increase of κ above the plateau is due to the hopping transport of strongly localized modes. Further, it was suggested that the quasiperiodicity affects the thermal conductivity in the same way as a simple aperiodicity.[39] Similar attempt, hopping conductivity, has been made by Janot to explain the monotonic increase of thermal conductivity of quasicrystal above 100 K.[49] It is noticed that the $\kappa(T)$ characteristics of the presently investigated Cu-Zr-Ti BMG show a significant resemblance to our previous report on Ag-In-Yb quasicrystal, except the presence of a clear maximum around 30 K was observed in quasicrystals.[44] Such an observation is in good agreement with the earlier reports that the low-temperature maximum in κ can be reduced by disorder or defects, or both.[45,46]

IV. CONCLUSIONS

In conclusion, we report the low temperature transport properties of BMG alloys $Cu_{64}Zr_{28.5}Ti_{7.5}$, $Cu_{62.3}Zr_{23.7}Ti_{10}$, $Cu_{60.6}Zr_{26.9}Ti_{12.5}$, and $Cu_{58.8}Zr_{26.2}Ti_{15}$ in the temperature range 10–300 K. All the presently investigated compositions show weak temperature-dependent variation in electrical resistivity with negative temperature coefficient. The negative TCR is most likely due to the weak localization effect, as evident by the magnetoresistivity measurements. It was noticed that the substitution of Ti enhances the disorder and hence ρ increases with increasing Ti concentrations. On the other hand, the temperature-dependent characteristics of thermopower and thermal conductivity were found to be almost identical, irrespective of variations in the compositions. It is found that the phonon contribution to the total thermal conductivity is nearly equal to the electronic contribution in the presently investigated Cu-Ti-Zr alloys, which is in contrast to the general impression that the glassy material generally has lower lattice thermal conductivity. The possible explanation for near identical characteristics in thermopower is that the Fermi level is masked from the changes in the electronic structure or in other words, the resonant levels of Cu and Ti/Zr are well above or below the Fermi level, respectively.

The thermopower of the Cu-Ti-Zr BMG alloys shows two main features that represent a deviation from the expected linear increase of diffusion thermopower with temperature. First, there is a knee (or change in slope) in the thermopower around 120 K that we have accounted for by the expected enhancement of metallic diffusion thermopower by the electron-phonon interaction at low temperatures. Second, a plateau is seen below 50 K that we have shown corresponds to an additional thermopower peak of approximately 0.17 μV/K centered at 22 K. This peak could be a remanent phonon drag peak, but it does correlate with the plateau seen in the thermal conductivity below 50 K and could well be associated with low-energy excitations in glasses.

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*Electronic address: ykkuo@mail.ndhu.edu.tw

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