PHYSICAL REVIEW B VOLUME 33, NUMBER 2 15 JANUARY 1986

Electronic-transport properties of amorphous Cu-Ti films

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(Received 5 August 1985)

We have measured the thermopower and resistivity of vapor-deposited amorphous Cu-Ti films of widely varying composition, at temperatures from 4.2 to 300 K. An explanation of the temperature variation in resistivity, but *not* in thermopower, requires a mechanism additional to the temperature dependence of the structure factor in the Ziman-Faber model. The magnitude of the resistivity, its temperature variation, and the thermopower are all larger in a $Cu_{47}Ti_{53}$ film than in corresponding melt-spun metallic glass ribbons, indicating greater disorder scattering, but differences between Cu-rich films and ribbons are more complex.

I. INTRODUCTION

Amorphous metal alloys can be made by a variety of different methods,[1] starting from the liquid phase (rapid quenching), vapor phase (deposition on a substrate), solid phase (irradiation or particle bombardment of a crystal), or liquid solution (chemical or electrodeposition). Although the properties of amorphous systems prepared by the various methods are similar, some differences exist—for example, amorphous alloys produced by irradiation methods lack the high degree of short-range order typically present in liquid-quenched systems.[1]

The electronic properties of amorphous metal ribbons prepared by rapid quenching from the melt (for which the term "metallic glass" is usually reserved) have been extensively studied.[2,3] Far fewer data, however, are available for vapor-deposited films. Naugle[2] points out that measurements of electron-transport properties in amorphous transition-metal systems (which typically possess high resistivities) have been almost entirely confined to metallic glasses. Thermopower measurements are particularly sparse for all classes of amorphous films. Only two previous groups (to our knowledge) have measured the thermopower of vapor-deposited films: Wiebking[4] measured the thermopower of Pb, Sn, Cu, Au, and dilute *Au*Fe films formed by vapor deposition at helium temperatures, and Korn and Mürer[5] measured the resistivity and thermopower of vapor-deposited Sn-Cu films. All these systems have relatively low resistivity, and irreversible annealing effects occur for most samples well before room temperature is reached, particularly, of course, for the pure-metal samples of Wiebking. Cote and Meisel[6] and Carini *et al.*[7] have, however, measured the thermopower of *electrodeposited* or *chemically deposited* NiP films, an example of the transition-metal—metalloid class of amorphous alloys, which have an intermediate resistivity from 100 to 165 $\mu\Omega$ cm.

In this paper we report the first measurements of thermopower in high-resistivity vapor-deposited films (no measurements of thermopower of *any* films with resistivity greater than 170 $\mu\Omega$ cm are listed by Mizutani[3] in his recent comprehensive review). High resistivity amorphous systems are of particular interest because their resistivity typically *decreases* as temperature increases in accordance with the Mooij correlation.[8] The explanation of this decrease is still controversial, the main contenders being temperature dependence of the structure factor in the Ziman-Faber model[9-12] or incipient localization.[13-15] We search in this paper for systematic differences in electronic transport properties between vapor-deposited films and metallic glass ribbons for a high-resistivity system involving only transition or noble metals, including measurements of thermopower, which is related to the energy dependence of resistivity. We chose the system CuTi since:

(i) Its resistivity[16] in ribbon form is well above the Mooij correlation resistivity of about 150 $\mu\Omega$ cm for which negative temperature coefficients of resistivity are expected.

(ii) Several investigators[16-20] have made corresponding measurements on metallic glass ribbons for comparison.

(iii) It can be obtained in amorphous form over a wide range of compositions and is stable at room temperature.

(iv) No complications are expected from magnetic effects, which can produce strongly energy-dependent scattering (and hence complicated thermopower and resistivity behavior).

II. EXPERIMENTAL DETAILS

The films were prepared by co-evaporation of Cu from a molybdenum boat and Ti from an electron-gun-heated source in a vacuum chamber with a base pressure of 3×10^{-9} Torr, rising to 10^{-7} Torr during the evaporation. The evaporation rates were monitored and controlled using quartz crystal microbalances, thus ensuring compositional uniformity through the entire film thickness. Samples were condensed on to room-temperature glass substrates ($50\times10\times1$ mm^3) at rates of between 4 and 12 Å s^{-1}. The compositions of the films were measured to within 2 at. % by x-ray fluorescence. Sample thicknesses were measured with a Varian Å scope. During each evaporation we deposited films on thin polythene sheet, and x-ray scattering was performed on these to establish the amorphous character of our samples.

Thermopower measurements were made using manganin reference leads that were calibrated against 99.999% pure Pb using the absolute thermopower of Pb determined by Roberts.[21,22] Electrical contact to the films was accomplished by soldering the manganin leads onto the inside of phosphor bronze clips using nonsuperconducting solder, so that the reference leads contacted the film when the assembly was clipped onto the sample. The thermometers, carbon resistors for $T<40$ K and transistors for $T>30$ K, were glued directly to the sample-substrate assembly. Data were taken using the differential method, introducing a small temperature difference ΔT across the sample and measuring the resultant change in voltage across the room temperature ends of the manganin reference leads. This was done for a series of values of ΔT at each temperature to ensure that the voltage was linear in temperature gradient.

Resistivity measurements were made with a standard four-terminal method, using spring loaded contacts. The contact geometry could not be very accurately determined, and this contributed, along with the uncertain average film thickness determinations, to the approximately 15% uncertainty in the form factor (length/cross-sectional area) and in the absolute value of the resistivities. The geometry was entirely stable, however, and as a consequence the accuracy of the temperature dependence of the resistivity, $\rho(T)/\rho(0)$, was limited only by the accuracy of the voltage and current measurements.

Both the thermopower and the resistivity measurements required reliable thermometers. For these we have used a variety of items (transistors and carbon, copper and Ge resistors) whose calibrations were repeatedly checked against each other and against a *Rh*Fe thermometer which had been calibrated by Cryogenic Calibration Ltd.

III. RESULTS AND DISCUSSION

Typical results for our amorphous Cu-Ti films are shown in Fig. 1 (thermopower) and Fig. 2 (resistivity), compared to previous results for melt-quenched ribbons of similar composition. Our data, and previous data for Cu-Ti ribbons, are summarized in Fig. 3. We now discuss these data in turn.

A. Thermopower

One of the most striking aspects of the thermopower results in Figs. 1 and 3 is the different size of some of our film thermopowers compared to those for the corresponding metallic glass ribbons. Absolute thermopower can be determined with good accuracy even for films, since unlike resistivity it is not dependent on accurate measurement of sample dimensions. A check of our calibration is provided by our results for one of the Leeds Cu-Zr ribbons,[16] which duplicate those of Gallagher and Greig[16] to within ±0.05 μV/K. This uncertainty is more than an order of magnitude smaller than the difference between the film and ribbon thermopowers in Fig. 1. In addition, the film samples were cycled between room temperature and liquid-nitrogen temperature several times with no detectable change in the thermopower, indicating the absence of relaxation or annealing effects. The difference between the film and ribbon samples is therefore real and reproducible. Possible explanations are discussed below.

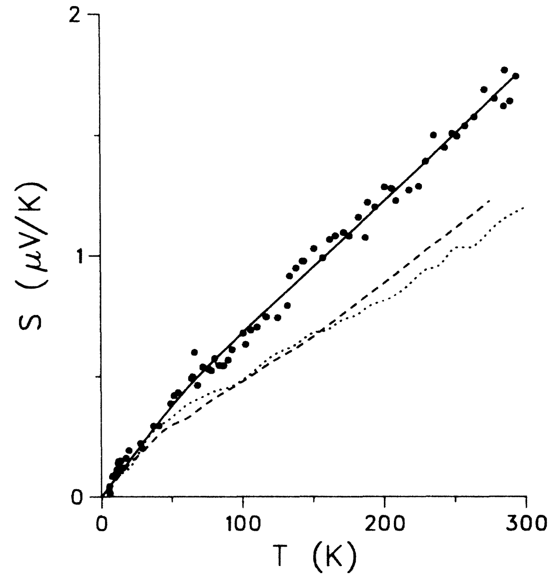

FIG. 1. Thermopower measurements on an amorphous $Cu_{47}Ti_{53}$ film (solid circles), fitted to our theoretical Eq. (1), with $x_b=5.8$ nV/K^2 and $\eta\lambda_{ph}(0)=0.3$ (solid line). Also shown for comparison are measurements for melt-quenched $Cu_{50}Ti_{50}$ metallic glass ribbons by Fritsch *et al.* (Ref. 19, dotted line) and Gallagher and Hickey (Ref. 20, dashed line).

Another significant aspect of the thermopower data is the lack of any unusual temperature dependence. As shown in Fig. 1, the overall behavior is approximately linear except for a knee at about 50 K. This knee is a characteristic feature of metallic glass thermopower, and there is strong evidence[16,20,23] that it is caused by electron-phonon enhancement of the bare diffusion ther-

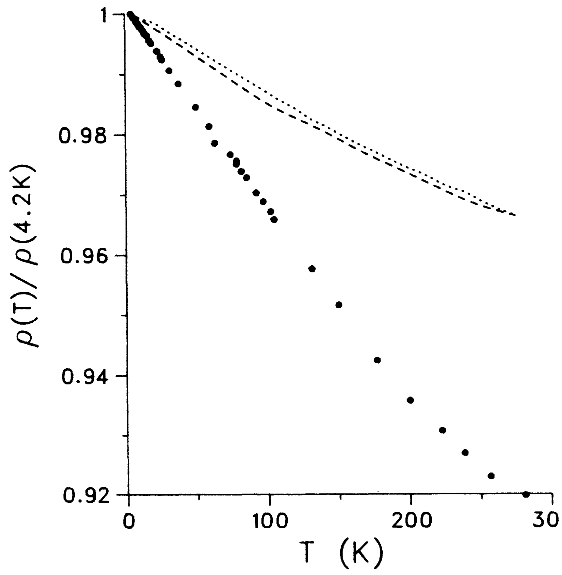

FIG. 2. Resistivity measurements on amorphous $Cu_{47}Ti_{53}$ film (dots), compared to data on melt-quenched $Cu_{50}Ti_{50}$ ribbons from Mizutani *et al.* (Ref. 17, dotted line) and Howson (Ref. 18, dashed line).

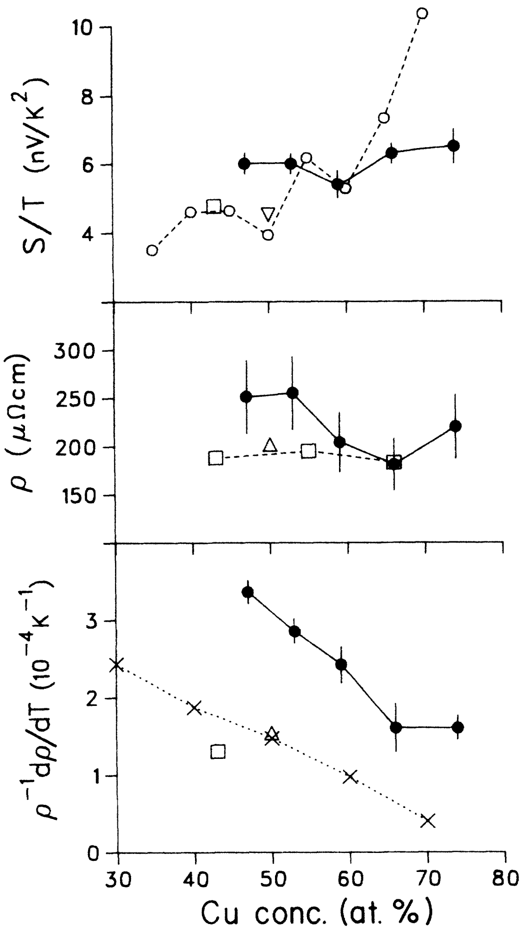

FIG. 3. Our data on amorphous Cu-Ti films (solid circles) for thermopower parameter S/T at 250 K, resistivity ρ at 4.2 K, and temperature coefficient of resistivity $\rho^{-1}d\rho/dT$ in the range 20—100 K. Other points are for melt-quenched Cu-Ti ribbons, from Ref. 16 (squares), Ref. 17 (crosses), Ref. 18 (upright triangles), Ref. 19 (open circles), and Ref. 20 (inverted triangles). Values for S/T from Ref. 19 are for a temperature of 300 K rather than 250 K, but the difference is small.

mopower, phonon-drag effects being essentially absent in amorphous systems.[24] The metallic diffusion thermopower in the presence of large disorder scattering is calculated[23] as

$$S=x_bT[1+\eta\lambda^s_{\text{ph}}(T)] \ . \tag{1}$$

Here, T is temperature, η is a constant, and x_b is the bare thermopower parameter given by the Mott formula in terms of the resistivity $\rho(\epsilon)$, where ϵ is the bare electron energy (i.e., excluding electron-phonon enhancement), by

$$x_b=\frac{\pi^2k_B^2}{3\,|e\,|}\left.\frac{\partial\ln\rho(\epsilon)}{\partial\epsilon}\right|_{\epsilon=\epsilon_F} . \tag{2}$$

In this equation k_B is Boltzmann's constant, e is electronic charge, ϵ_F is the Fermi-energy, and $\lambda^s_{\text{ph}}(T)$ is the temperature-dependent electron-phonon enhancement of thermopower defined by Kaiser,[23] which equals the usual mass enhancement $\lambda_{\text{ph}}(0)$ at zero temperature. This expression includes velocity and relaxation-time renormalization effects, and the contribution from higher-order Feynman diagrams (the Neilsen-Taylor effect), which for amorphous systems give the same temperature dependence[23] as the energy renormalization due to the electron-phonon interaction. The effect of the presence of these additional terms is to change the value of the constant η from the value of unity corresponding to the energy-renormalization effect alone.

We have performed detailed fits of our data to the theoretical expression (1), with fitting parameters x_b and $\eta\lambda_{\text{ph}}(0)$, using a Debye model (with an experimental Debye temperature[25] of 340 K) to calculate the temperature dependence of the electron-phonon enhancement $\lambda^s_{\text{ph}}(T)$; this aspect of our data has already been discussed in a brief report.[26] It can be seen from Fig. 1 that the enhancement effect is rather similar in the ribbon samples.[19,20] Fritsch *et al.*[19] see the same decrease of the enhancement toward the Cu-rich end of the compositional range that we found,[26] and that is obviously consistent with the smaller electron-phonon enhancement in Cu, but their values for the enhancement are all significantly larger than ours. Since each group[19,20,26] uses different methods for estimating low-temperature enhancement, we have fitted the ribbon data in Fig. 1 to the full theoretical expression of Eq. (1) (with Debye temperature 340 K and equal weighting of S values) as a check. Our fits give $\eta\lambda_{\text{ph}}(0)=0.35$ for the data of Gallagher and Hickey[20] (compared to their quoted value of 0.4), and 0.65 for the $\text{Cu}_{50}\text{Ti}_{50}$ ribbon of Fritsch *et al.* (compared to their value of 0.8), essentially confirming the analyses of these authors. Our effective enhancements[26] of 0.30 for the $\text{Cu}_{47}\text{Ti}_{53}$ film and 0.35 for the $\text{Cu}_{53}\text{Ti}_{47}$ film agree with that for the data of Gallagher and Hickey, so it is not clear that the systematic difference between our results and those of Fritsch *et al.*[19] is between films and ribbons. We believe the uncertainty in all these determinations of effective enhancement is at least ±0.1 and is very sensitive to small uncertainties in the experimental thermopower at low temperatures.

We examine now the implications of the thermopower data for theoretical models. In the Ziman-Faber model, the bare thermopower $S_b=x_bT$ is predicted to be linear in T at low temperatures, with x_b given[27] by substituting in Eq. (2) the expression for resistivity[9]

$$\rho(\epsilon)=\frac{12\pi\Omega}{e^2\hbar V_F^2}\int_0^1 d\left[\frac{K}{2K_F}\right]\left[\frac{K}{2K_F}\right]^3 a(K)\,|\,t(K)\,|^2 \ , \tag{3}$$

where Ω is the volume per atom, V_F the Fermi velocity, K_F the Fermi wave vector, K the scattering wave vector, $a(K)$ the resistivity structure factor, and $t(K)$ the single-site t matrix. We obtain[2]

$$x_b=\frac{\pi^2k_B^2}{3\,|e\,|\,\epsilon_F}\xi \ , \tag{4}$$

where

$$\xi=-3+2q+r/2 \ , \tag{5}$$

$$q=a(2K_F)\,|\,t(2K_F)\,|^2/\langle a(K)\,|\,t(K)\,|^2\rangle \ , \tag{6}$$

$$r=K_F\left\langle \frac{\partial}{\partial K}\,|t(K)|^2a(K)\Big|_{K=K_F}\right\rangle \Big/ \langle a(K)\,|t(K)|^2\rangle \;, \tag{7}$$

and

$$\langle f(q)\rangle=4\int_0^1 d\left[\frac{K}{2K_F}\right]\left[\frac{K}{2K_F}\right]^3 f(K)\;. \tag{8}$$

For a binary alloy $a(K)\,|t(K)|^2$ should be replaced by a rather complicated expression involving the three partial structure factors.[2] When $2K_F$ is in the vicinity of the wave vector K_p of the first peak in the structure factor, we would obviously expect q to be considerably greater than unity, providing a mechanism for positive thermopowers in the Ziman-Faber model.[28] Such dominance of scattering by the first structure-factor peak would also, of course, give a high resistivity and negative temperature coefficient of resistivity. However, we would not expect the condition $2K_F \sim K_p$ to apply right across the wide concentration range investigated in this work. Further, as pointed out by Gallagher and Greig,[16] the energy dependence of the t matrix, represented by the term r in Eq. (5), can make a dominant contribution (but is rather difficult to estimate accurately). We *do* expect q and r to be largely independent of temperature, giving a linear bare thermopower as observed. Note, however, that the T dependence of the structure factor $a(K)$ could produce some T dependence in x_b as in the resistivity discussed below, which could lead to some curvature in the thermopower,[19,29] particularly at higher temperatures. Thus we conclude that the observed thermopower is at least qualitatively consistent with the Ziman-Faber model.

Turning to the differences between the film and ribbon thermopowers shown in Fig. 3, we note that the larger thermopower for our $Cu_{47}Ti_{53}$ film is consistent with the film resistivity and its temperature coefficient also being larger, since empirically systems with higher resistivity tend to have a positive thermopower[2]—this is the analog for thermopower of the Mooij correlation. This correlation is unlikely, however, to account for the large increase in thermopower for the Cu-rich ribbons[19] that is absent in our films. Fritsch *et al.*[19] suggested that their CuTi ribbon thermopowers showed evidence of varying short-range chemical order quenched in from the liquid. Such order will be reduced in vapor-deposited films, leading to different behavior of the thermopower, which can be very sensitive to structural differences, through the parameter r in Eq. (5). Increasingly localized Ti d levels lying above the Fermi energy would be expected to produce greater scattering of conduction electrons *above* the Fermi energy, giving a positive thermopower. The more random structure in the films could reduce the structure-related effects seen in the ribbons, giving a less variable thermopower.

The approximately linear T dependence of thermopower, except for the effect of the expected decay of electron-phonon enhancement, has implications for alternative models for transport properties. If significant conduction were occurring by thermally excited hopping, a nonlinear thermopower contribution (for example, $T^{1/2}$) might be expected.[30] No such contribution is seen. Even well into the metallic regime, weak localization effects (as well as electron-electron correlation effects at low temperatures) could influence conductivity.[15,18,31] The absence of any observed additional nonlinear thermopower effects implies one of two possibilities, between which we cannot at present decide.

(i) The thermopower due to incipient localization effects could be the same as that in their absence. For example, Ting *et al.*[32] have calculated that weak localization in their two-dimensional (2D) model makes no difference to thermopower (but electron-electron correlations do). If this were true in 3D, significant localization effects could be present in resistivity but not in thermopower. Note that a *linear* thermopower characteristic of weak localization different in size to that characteristic of disorder scattering would in general lead to a nonlinear total thermopower if the importance of weak localization effects decreased significantly as temperature increases.

(ii) The contribution of electron-electron correlation or weak localization effects to resistivity could be nonexistent in our temperature range, or sufficiently small that the thermopower remains that characteristic of the dominant static disorder scattering. Note that the relative accuracy of measurements of thermopower is far less than those of resistivity, especially at low temperatures where thermopower goes to zero. Hence changes in S/T of a few percent due to localization or electron-electron effects could escape detection or be masked by the electron-phonon enhancement knee, whereas similarly sized effects in resistivity might be clearly seen.

B. Resistivity

Figures 2 and 3 show that the temperature dependence of resistivity for our CuTi films follows the same trend as a function of concentration as that for CuTi ribbons[17] but is about twice the magnitude. This greatly increased T dependence makes an explanation of the resistivity within the extended Ziman-Faber model more unlikely, as we now show.

In a simplified treatment in the framework of the extended Ziman-Faber model, Cote and Meisel[9] obtained

$$\rho=(1-\gamma)\rho_{ip}+\rho_0 e^{-2[W^*(T)-W^*(0)]}\;, \tag{9}$$

where ρ_{ip} is the ideal phonon resistivity (for a large mean free path), ρ_0 is the residual resistivity (due to elastic scattering), γ is a coefficient representing the extent of the suppression of the electron-phonon scattering owing to the mean free path being shorter than typical phonon wavelengths, and $2W^*$ is an average Debye-Waller exponent[33]

$$2W^*(T)=\frac{3\hbar^2K^2}{Mk_BT_D}\left[\frac{T}{T_D}\right]^2\int_0^{T_D/T} x[(e^x-1)^{-1}+\tfrac{1}{2}]dx\;, \tag{10}$$

where M is atomic mass and T_D is the Debye temperature. We have inserted $W^*(0)$ in Eq. (9) since $W^*(T)$ does not go to zero as $T\rightarrow 0$, owing to the zero-point fluctuations.

For a very high resistivity system, Cote and Meisel[9] suggest that substantial suppression of the phonon resistivity term (which *increases* with T) could occur. If we take $\gamma=1$, corresponding to complete suppression, the T dependence of the resistivity is simply that of the average Debye-Waller factor. This T dependence represents the *maximum* decrease of resistivity in this model.

Equation (9) with $\gamma=1$ is equivalent to the expression of Nagel,[11] except that in Nagel's model only the part of the structure factor $a(2k_F)$ greater than unity decays with the Debye-Waller factor (only the structure factor at $2k_F$ is considered because backscattering is assumed to dominate):

$$\rho=\rho_0 a^{-1}(2k_F)\{1+[a(2K_F)-1]e^{-2[W^*(T)-W^*(0)]}\} . \quad (11)$$

For high-resistivity systems, $a(2K_F)$ will be considerably greater than unity, and again the decay of the Debye-Waller factor represents the *maximum* decrease of resistivity with temperature.

This Debye-Waller T dependence is shown in Fig. 4 for Cu-Ti, with $M=55\times$(proton mass), $T_D=340$ K, and $K\approx3$ Å^{-1}, the approximate location of the first peak in the structure factor. Also shown are our experimentally observed temperature dependences of conductivity (conductivity is used rather than resistivity for the discussion of localization below). It is clear that the Debye-Waller factor cannot account for all of the observed temperature dependence of conductivity or resistivity. Mizutani *et al.*[17] pointed out that their Cu-Ti ribbon resistivity at low temperatures disagreed with the T^2 law expected in the Cote and Meisel model;[9] our $Cu_{47}Ti_{53}$ film resistivity shows disagreement even at intermediate temperatures.

Unlike thermopower, therefore, the resistivity requires mechanisms beyond the usual Ziman-Faber model for the explanation of its T dependence. Our data in Fig. 4 show approximately linear behavior over the temperature range 10–100 K, with a slower variation at higher temperatures. This behavior is qualitatively similar to that of Howson[18] for his Cu-Ti ribbon and ascribed to localization effects (predicted to give a linear T dependence in conductivity at low temperatures, changing to $T^{1/2}$ at higher temperatures). However, the Debye-Waller factor decay of the elastic resistivity is also likely to be a partial cause of the observed T dependence at higher temperatures.

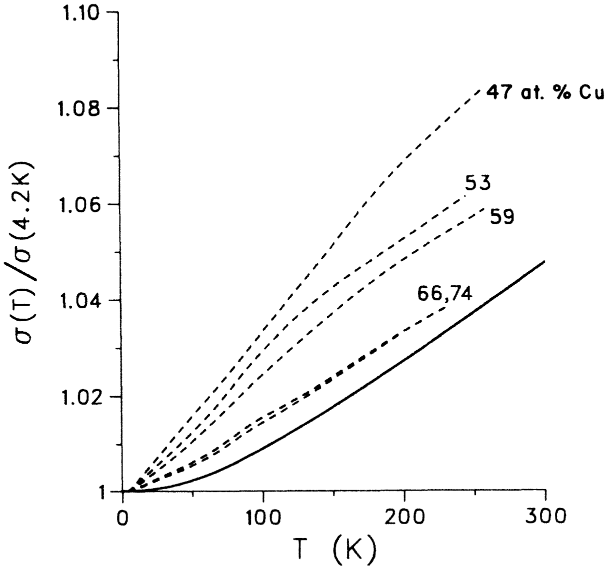

FIG. 4. Conductivity of our amorphous Cu-Ti films (relative to its value at a temperature of 4.2 K), with Cu concentration indicated beside each curve. For comparison, the maximum conductivity change expected from decay of the Debye-Waller factor in the extended Ziman model, as given by Eq. (11) with $a(2K_F)\gg1$, is also shown (solid line).

As mentioned above, the accuracy of absolute resistivity values for the films is not good. Nevertheless, the data in Fig. 3 indicate that our films tend to have higher resistivity than the corresponding ribbons (the resistivity values at 300 K of 206–186 $\mu\Omega$ cm quoted by Mizutani *et al.*[17] for Cu-Ti ribbons containing 40% to 60% Cu, and of 200–176 $\mu\Omega$ cm for those of Fritsch *et al.*[19] with 35% to 60% Cu, are clearly consistent with the low-temperature resistivity values for the other ribbons shown in Fig. 3). The larger negative temperature coefficients of resistivity for our films are therefore consistent with the Mooij correlation.[8]

For a low-resistivity amorphous system, Nguyen *et al.*[34] found a greater resistivity in vapor-deposited Mg-Zn films compared to melt-quenched glasses, but a smaller negative temperature coefficient of resistivity. For vapor-deposited Ag-Cu-Ge films, Mizutani and Yoshida[35] found no significant difference in resistivity magnitude or temperature coefficient compared to ribbons.

IV. CONCLUSION

We have found that the magnitude of the resistivity ρ in our vapor-deposited $Cu_{47}Ti_{53}$ and $Cu_{53}Ti_{47}$ films is higher than that in corresponding glassy metal ribbons. Consistent with the Mooij correlation and its thermopower analog, the negative temperature coefficient of resistivity and the thermopower magnitude are also larger. Thus all three observations are consistent with greater disorder scattering in the films. Thermopower differences for the Cu-rich films and ribbons, however, are not accounted for by the Mooij correlation.

We note that Zweck and Hoffmann[36] suggested their electron scattering and high-resolution electron microscopy observations on amorphous FeB ribbons and sputtered films could actually be explained in terms of a microcrystallite model, with crystallite diameter of about 2 nm. Clearly if such strong short-range order is present in other amorphous systems, significant variations in the size and character of ordered regions might occur for different methods of preparation. For example, atom-by-atom accretion in vapor deposition could give a more random structure (and so higher resistivity) than quenching of a liquid which already contains substantial short-range order.[1]

The greater temperature dependence of resistivity for our films emphasizes that the decay of disorder resistivity via the Debye-Waller factor in the extended Ziman model is inadequate to account for this temperature dependence, particularly at lower temperatures. The approximately linear T dependence from 10 to 100 K is similar to that seen by Howson in a $Cu_{50}Ti_{50}$ ribbon and ascribed to incipient localization effects. However, the thermopower is approximately linear in temperature between 5 and 300 K except for the electron-phonon enhancement effect at lower temperatures, and so is qualitatively consistent with the extended Ziman model, providing no evidence for localization effects.

ACKNOWLEDGMENTS

We thank Dr. Neil Whitehead of the Institute of Nuclear Sciences, Department of Scientific and Industrial Research, Lower Hutt, for the x-ray fluorescence measurements, Dr. Bryan Gallagher and Dr. Davor Pavuna for their collaboration, and the New Zealand University Grants Committee and the Victoria University of Wellington Internal Research Committee for financial support. One of us (K.D.D.R.) acknowledges partial support from the New Zealand Commonwealth.

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