Nonuniversality of the Mooij Correlation—the Temperature Coefficient of Electrical Resistivity of Disordered Metals

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(Received 27 August 1986)

By using compiled, updated experimental data, we have demonstrated that the correlation between the temperature coefficient of electrical resistivity α and the resistivity ρ for disordered metals at room temperature is not universal. The origin of the nonuniversality of the Mooij correlation stems mostly from a competition between the quantum mechanical effects of incipient localization and the classical Boltzmann electron transport. Results of a numerical analysis based on localization indicate that there is a unique and monotonic correlation between α and ρ for a specific disordered metallic system, in accord with the essential features of our new Mooij (α vs ρ) plot.

PACS numbers: 72.15.Eb, 71.55.Jv

More than a decade ago, Mooij found that, at room temperature, the sign and the size of the temperature coefficient of electrical resistivity [$\mathrm{TCR} \equiv \alpha(\rho) \equiv \rho^{-1} \times \partial\rho/\partial T$] correlate well with the magnitude of the resistivity (ρ) in many disordered metallic systems.[1] Furthermore, Mooij observed that α changes sign in a relatively narrow range of resistivity (i.e., the critical resistivity for which $\alpha = 0$, $\rho_c \cong 100$–$150\ \mu\Omega$ cm). In the literature, a resistivity value of $150\ \mu\Omega$ cm has often been given fundamental significance in the sense that it serves as a universal boundary which divides the positive and negative TCR's. In this Letter, I will demonstrate that ρ_c is not universal and, in fact, it can have values ranging from 30 to as high as $400\ \mu\Omega$ cm and above (Fig. 1). Also, I will use the concept of electron localization to understand the nonuniversality of Mooij correlation. I will also show that there is a unique and monotonic correlation between α and ρ for a specific disordered metallic system. This correlation may be very useful in extracting valuable information about various inelastic electron scattering processes, the electron-phonon mean free path, for example.

The data collected in the original Mooij (α vs ρ) plot were mostly for crystalline disordered metallic systems (122 points), and there were only 11 data points for amorphous metals. In this work, I have compiled more than 500 data points collected from the literature.[2] The results for $-5.0 \leq \alpha \leq +4.0$ (in units of $10^{-4}\ \mathrm{K}^{-1}$) as a function of ρ are presented in Fig. 1. For comparison, solid lines outline the region where the original Mooij data points were distributed. Several salient features emerge from Fig. 1. Many data points lie outside of the Mooij region which suggests that the original, approximately linear correlation between α and ρ is not an accurate summary of the experimental data. The magnitude of the critical resistivity ρ_c depends very much on the individual material, and can vary from 30 to $400\ \mu\Omega$ cm. Clearly, the data show that no fundamental significance should be attached to the resistivity value of $150\ \mu\Omega$ cm. For metals with room-temperature resistivity $\rho \lesssim 200\ \mu\Omega$ cm, it appears that there is no tangible correlation between α and ρ. The fact that many relatively low-resistivity ($\rho \sim 50\ \mu\Omega$ cm) amorphous metals are characterized by relatively large negative TCR's shows that resistivity alone cannot determine the sign of TCR in metals. For $\rho > 200\ \mu\Omega$ cm, the TCR tends to be more negative with increasing ρ. The data presented in Fig. 1 encompass those of a wide variety of disordered metals. The negative TCR can be found in the crystalline as well as in the amorphous state, in bulk samples as well as in thin films. Apparently, no particular kind of disorder favors the occurrence of negative TCR's. In short, the experimental data compiled in

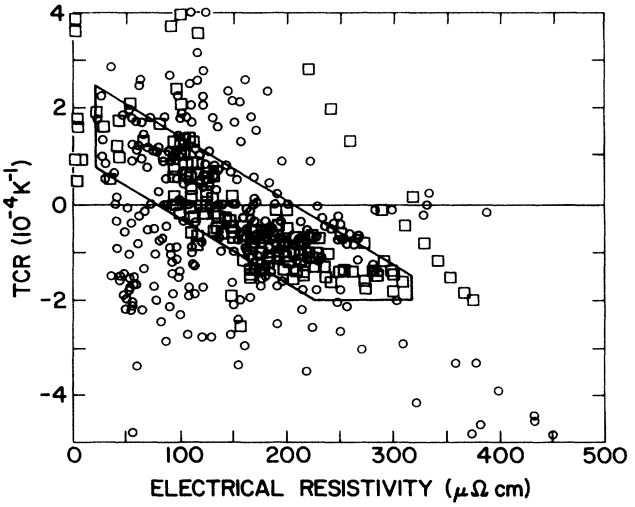

FIG. 1. TCR vs electrical resistivity for various crystalline disordered metallic conductors (open boxes) and amorphous metals (open circles) at room temperature. The solid lines were used to outline the region where the original Mooij data points were distributed.

Fig. 1 suggest that the correlation between α and ρ in disordered metals is nonuniversal, contrary to the original Mooij suggestion.

In this Letter, I propose to use the concept of electron localization to study the resistivity dependence of α for various disordered metallic systems. The Mooij correlation has been discussed qualitatively by several authors in terms of localization, but the issue of nonuniversality has never been considered.[3-5] For a critical review on this topic, the reader is referred to the article by Lee and Ramakrishnan.[6] Following Kaveh and Mott,[5] we begin our discussion of α in disordered metals with quantum corrections to the Boltzmann conductivity $\sigma_B(T)$ for the three-dimensional case:

$$\delta\sigma(T)=\sigma_B(T)-\sigma(T)=\frac{e^2}{\pi^2\hbar}\left[\frac{1}{L_i(T)}-\frac{1}{l_e}\right], \quad (1)$$

where l_e is the elastic mean free path and L_i the inelastic diffusion length as defined by $L_i=(\frac{1}{3}l_e l_i)^{1/2}$, with l_i the inelastic mean free path. From Eq. (1), we obtain

$$\sigma(T)=\sigma_B(0)\left[1-\frac{3}{(k_F l_e)^2}\right]+\frac{e^2}{\pi^2\hbar}\left[\frac{1}{L_i(t)}-\frac{(k_F l_e)^2}{3l_i(T)}\right]. \quad (2)$$

In arriving at Eq. (2), I have assumed that $l_e \ll l_i$, a condition satisfied even at room temperature for most highly disordered metals as a result of the fact that l_e is of the order of interatomic distances in such materials. The first term in Eq. (2) represents the zero-temperature, quantum-corrected conductivity $\sigma(0)$ due to localization; the second term describes the competition between the degradation of the quantum-interference effects as a result of inelastic scattering and the conventional thermal excitation of various inelastic processes. Obviously, the balance between these competing effects on the current-transport electrons determines the sign and the size of α as a function of temperature. Before we use Eq. (2) to derive an expression for $\alpha(T)$, the following experimental facts should be emphasized:

(i) For most disordered metals, the temperature-dependent part of the conductivity in the temperature range of 0–300 K is generally 10% or less of $\sigma(0)$.

(ii) The value of σ for highly disordered metallic conductors is about 5–20 times higher than that of the Mott minimum conductivity. This is consistent with the fact that $k_F l_e$ for most amorphous metals is about 4–10 which is still significantly above the Joffe-Regel criterion for a metal-insulator transition.[6] We expect, therefore, that the weak-localization assumption on which Eq. (1) is based is valid, and it is reasonable to approximate $\alpha(T)$ for disordered metallic conductors by the following expressions:

$$\alpha(T)\equiv-\frac{1}{\sigma(T)}\frac{\partial\sigma(T)}{\partial T}\cong-\frac{1}{\sigma_B(0)}\frac{\partial\sigma(T)}{\partial T}. \quad (3)$$

From Eq. (2), it is straightforward to obtain the following result:

$$\alpha(T)=-\frac{P}{T}\left[\frac{3\sqrt{3}}{2}l_i^{-1/2}l_e^{-3/2}k_F^{-2}-l_i^{-1}l_e\right]\times\left[1-\frac{l_e}{l_i}+\frac{3l_e}{(k_F l_e)^2}\left(\frac{1}{L_i}-\frac{1}{l_e}\right)\right]^{-1}. \quad (4)$$

As usual,[6] the temperature dependence of l_i is expressed by $l_i(T)=AT^{-P}$, where A is a constant, and the value of P can vary with temperature (e.g., $P=2$–5 for $T<40$ K, $P=1$ for $T>200$ K).

From Eq. (4), the condition for $\alpha=0$ at $T=T_c$ can be readily obtained:

$$l_i(T_c)/l_e=\tfrac{4}{27}(k_F l_e)^4. \quad (5)$$

By applying this condition to Eq. (2), we get an expression for the critical resistivity ρ_c^{3D} for the three-dimensional case:

$$\rho_c^{3D}=\rho_B(0)\left[1-\frac{3}{(k_F l_e)^2}+\frac{6.75}{(k_F l_e)^4}\right]^{-1}, \quad (6)$$

where $\rho_B(0)=3\pi^2\hbar/e^2k_F^2l_e$. Equation (6) clearly indicates that the crossover point, ρ_c, in the α-vs-ρ plot is not universal and depends independently on k_F and l_e. In other words, ρ_c depends on the material characteris-

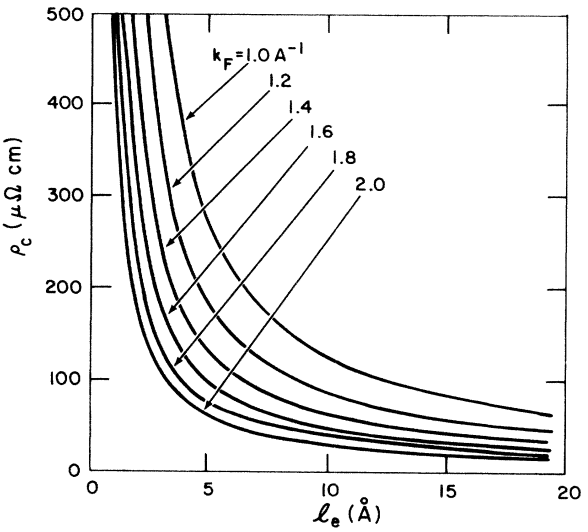

FIG. 2. The critical resistivity ρ_c as a function of l_e and k_F, based on Eq. (6).

tics (k_F) and its degree of disorder l_e.

According to Eq. (6), a plot of ρ_c vs l_e as a function of k_F is presented in Fig. 2. From this figure, one can understand quantitatively the nonuniversality of the Mooij correlation as depicted in Fig. 1. We recall the facts that the value of k_F for most metallic systems lies in the range of $1\ \text{Å}^{-1} < k_F < 2\ \text{Å}^{-1}$ and the elastic mean free path l_e of the conduction electrons in highly disordered systems cannot be much less than 2–3 Å. These experimental facts suggest a range of values for the crossover resistivity in disordered metals (i.e., 30–500 $\mu\Omega$ cm), which is in excellent agreement with the experimental data as shown in Fig. 1. In particular, the preponderance of ρ_c data around 150 $\mu\Omega$ cm (see Fig. 1) is merely a manifestation of the fact that many, if not most, disordered metallic conductors are characterized by an elastic mean free path l_e of 5 ± 2 Å and a Fermi wave vector k_F of $1.5 \pm 0.3\ \text{Å}^{-1}$. For certain low-electron-density systems[6] ($k_F < 1\ \text{Å}^{-1}$) such as Si:P, Eq. (6), however, predicts a value for ρ_c about 10^3 $\mu\Omega$ cm and higher.

For the low-dimensional cases, the conditions for $\alpha = 0$ and the formula for ρ_c can be obtained similarly as for the three-dimensional case. The results are listed as follows:

2D case:—For sample thickness $t < L_i^{2D}$, and $L_i^{2D} = \frac{1}{2}\sqrt{2}(l_e l_i)^{1/2}$,

$$l_i/l_e = \pi(k_F l_e) \text{ for } \alpha = 0, \tag{7}$$

and

$$\rho_c^{2D} = \frac{2\pi\hbar}{e^2}\{k_F l_e - \pi^{-1}[1 + \ln(\tfrac{1}{2}\pi k_F l_e)]\}^{-1}. \tag{8}$$

1D case:—

$$l_i/l_e = (4\pi^2)^{2/3} \text{ for } \alpha = 0, \tag{9}$$

$$\rho_c^{1D} = 0.63\pi\hbar/e^2 l_e. \tag{10}$$

We notice that, in the 2D case, ρ_c^{2D} depends only on $k_F l_e$ as a product and hence is material independent. In this sense, ρ_c^{2D} is universal for a given amount of disorder provided that the electron-interaction effects are relatively small as compared with that of localization. In reality, however, unlike in the 3D case, the electron-electron interaction usually cannot be ignored and the universality as suggested by Eq. (8) cannot be observed experimentally. The 1D case is clearly nonuniversal because ρ_c^{1D} depends on a material-dependent parameter l_e. The cases of mixed dimensions (e.g., classically 3D and quantum mechanically 2D) will be treated in a future publication.

Now back to α as a function of ρ for 3D disordered metals; one can study $\alpha(\rho)$ numerically for a given temperature by using both Eqs. (2) and (4). As can be seen from these equations, $\alpha(\rho)$ depends only on k_F and $l_i(T)$. If k_F is kept constant, the results of a numerical analysis show that α always decreases linearly with increasing ρ for $\rho \gtrsim \rho_c$. For $\rho < \rho_c$, α increases rapidly with decreasing ρ. The value of ρ_c, of course, is determined by Eq. (6) as discussed previously. The details of such a numerical study will be published later. As an example, a comparison between the theory and an experiment is shown in Fig. 3. The experimental data are taken from the work of Dynes, Rowell, and Schmidt[7] on the electrical resistivity of $LuRh_4B_4$ films (3000 Å) as a function of temperature and disorder induced by 1.8-MeV α-particle irradiation. The data points (dots shown in Fig. 3) are the digitized values for α derived from the ρ-vs-T curves of Fig. 5 in Ref. 7. The theoretical curve is obtained on the assumption that $k_F = 1.7\ \text{Å}^{-1}$ and $A = 5000$ Å K [$l_i(T) = AT^{-1}$]. We note that $k_F = 1.7\ \text{Å}^{-1}$ corresponds to a free-electron Fermi level of 11 eV, as suggested by a self-consistent energy-band calculation[8] on a similar ternary boride $ErRh_4B_4$. Also, I should point out that the only fitting parameter $A = 5000$ Å K corresponds to a reasonable value ~ 15 Å for the inelastic mean free path at room temperature. As indicated clearly in Fig. 3, the essential characteristics of the experimental data α as a function of disorder ρ can be described by the weak-localization model.

The agreement between the experimental data and numerical results probably could be improved if the effects of electron-electron interaction and spin-orbit scattering were included in the numerical analysis. Furthermore, we should take into account the possibility that inelastic scattering can be a function of the amount of disorder[9] in the sample. By and large, the evidence presented so far strongly suggests that electron localization plays a significant role in determining the sign as well as the magnitude of TCR in 3D disor-

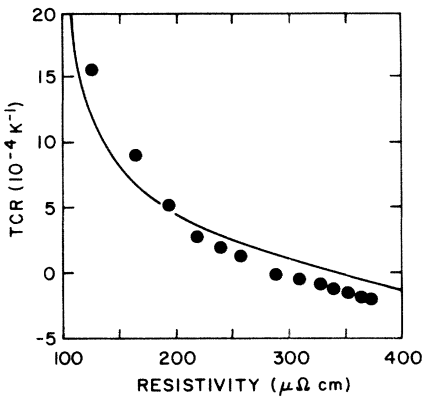

FIG. 3. TCR as a function of electrical resistivity at room temperature. The dots are data points and are the digitized values for TCR's derived from the ρ-vs-T curves of Fig. 5 in Ref. 7. The solid curve is obtained with a numerical calculation based on Eqs. (2) and (4), on the assumption that $k_F = 1.7\ \text{Å}^{-1}$, $l_i = AT^{-1}$, and $A = 5000$ Å K.

dered conductors even at room temperature. In addition to the temperature-dependent resistivity, numerous measurements[6] of magnetoconductivity, and of upper critical field in certain cases, have given strong supporting evidence for the importance of localization effects in 3D disordered metals at low temperatures ($T < 4$ K). For the room-temperature case, a magnetic field H of the order of 100 T is needed to observe a positive magnetoconductivity effect arising from the delocalization process by a magnetic field. This is best understood by comparing the magnetic and thermal (inelastic) dephasing length scales [$L_M = (\hbar/2eH)^{1/2}$, and $L_i(T)$ as defined earlier]. If localization is viewed simple-mindedly as an interference effect arising from electron coherent backscattering, a magnetic field of H can weaken the localization significantly only if the condition of $L_M < L_i(T)$ is satisfied. For example, to delocalize the highly disordered α-particle–irradiated films of $LuRh_4B_4$ ($L_i \cong 10$ Å at room temperature as inferred from the data fitting shown in Fig. 3), one would need a magnetic field of about 600 T, a magnetic field strength extremely difficult if not impossible to achieve experimentally. This is in accord with the fact that there has been no report of negative magnetoresistance as an evidence of localization for 3D disordered metals at room temperature. It, nevertheless, underscores the relative ease and importance of studying the temperature dependence of electrical resistivity as a function of disorder. In this sense, a numerical analysis based on Eqs. (2) and (4) represents a simple and viable way of probing the interplay between localization and inelastic scattering processes such as electron-phonon interaction.

In conclusion, I have demonstrated for the first time that the correlation between TCR and resistivity is nonuniversal and is more complex than the original Mooij correlation. The origin of this nonuniversality can be understood quantitatively in terms of a competition between incipient localization and the conventional Boltzmann electron transport at room temperature. By taking into account the quantum corrections to the Boltzmann conductivity, I have shown that the critical resistivity depends on Fermi level and disorder for 3D conductors and can have values ranging from 30 to about 400 $\mu\Omega$ cm for various disordered metals, in excellent agreement with the experimental data. By using numerical analysis, I also show that for a specific disordered metallic system, such as α-particle–irradiated $LuRh_4B_4$, there is a unique and monotonic correlation between α and ρ which can explain the essential features of the new Mooij (α vs ρ) plot.

I would like to thank C. C. Chi, K. Kelton, D. H. Lee, P. A. Lee, Y. Imry, and P. Santhanam for many useful discussions, and R. C. Dynes for providing a copy of Fig. 5 in Ref. 7 and for helpful comments.

[1]J. H. Mooij, Phys. Status Solidi A **17**, 521 (1973).

[2]The data points shown in Fig. 1 are taken from Ref. 1, U. Mizutani, Prog. in Mater. Sci. **28**, 97 (1983), S. R. Nagel, Adv. in Chem. Phys. **51**, 227 (1982), D. G. Naugle, J. Phys. Chem. Solids **45**, 367 (1984), and many other papers on electrical resistivity in disordered metals. A detailed description of this data compilation will be published elsewhere.

[3]Y. Imry, Phys. Rev. Lett. **44**, 469 (1980).

[4]M. Jonson and S. M. Girvin, Phys. Rev. Lett. **43**, 1447 (1979).

[5]M. Kaveh and N. F. Mott, J. Phys. C **15**, L707 (1982).

[6]P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).

[7]R. C. Dynes, J. M. Rowell, and P. H. Schmidt, in *Ternary Superconductors,* edited by G. K. Shenoy, B. D. Dunlap, and F. Y. Fradin (North-Holland, Amsterdam, 1981), p. 169.

[8]T. Jarlborg, A. J. Freeman, and T. J. Watson-Yang, Phys. Rev. Lett. **39**, 1032 (1977).

[9]Albert Schmid, Z. Phys. **259**, 421 (1973).