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Localization in thin copper films

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Resistance measurements in zero magnetic field have been made on thin Cu films (30 to 500 Å) between 1 to 100 K with the goal of determining the relevant scattering times. The elastic scattering time τ_0 is limited by the thickness d of the samples and is well described by $\tau_0 = v_F/d$. With the assumption that the inelastic scattering time τ_i has a temperature dependence given by $\tau_i \propto T^{-P}$, then the high-temperature resistance data yielded $P = 1.9 \pm 0.3$. These measurements also demonstrate that the localization phenomena extend to temperatures much greater than the temperature $T_{\min}$ at which the resistance minimum occurs. At low temperatures in the thinnest films, deviations in the resistance from the logarithmic temperature dependence are observed; this effect is attributed to strong spin-orbit scattering which dominates the inelastic scattering at low temperatures ($\tau_{so} \leq \tau_i$).

Numerous authors have shown that in thin, two-dimensional, (2D) metallic films, all of the electronic states should become weakly localized resulting in a logarithmic increase in the resistance as the temperature is lowered.[1] At low temperatures where the phonon contribution to the conductance of the sample can be neglected, the conductance per square is predicted to be

$$\sigma(T) = \sigma_0 - \frac{\alpha e^2}{2\pi^2\hbar} \ln \frac{\tau_i(T)}{\tau_0} \ (\Box/\Omega) \ , \tag{1}$$

where $\sigma(T)$ is the conductance per square of the film, σ_0 is the residual conductance per square at $T + 0$, α is a constant describing the interaction between the electrons, τ_i is the inelastic scattering time which has a temperature dependence given by $\tau_i \propto T^{-P}$, τ_0 is the elastic scattering time, and $e^2/2\pi^2\hbar = 1.23 \times 10^{-5} \ \Box/\Omega$. The residual conductance per square, σ_0, is defined as

$$\sigma_0 = ne^2\tau_0 d/m^* \ . \tag{2}$$

In our copper films, the logarithmic contribution to the conductance per square is very small (owing to the prefactor $e^2/2\pi^2\hbar = 1.23 \times 10^{-5} \ \Box/\Omega$) when compared to the residual conductance σ_0; in our samples, σ_0 typically takes on a value of 10^{-1} to $10^{-2} \ \Box/\Omega$. Thus resolution of ppm in the resistance is required to observe localization effects in our films.

Thin films become two dimensional when the inelastic diffusion length L_i becomes equal to or greater than the film thickness d. The average length of the localized electronic state is given by L_i, where L_i is defined as

$$L_i(T) = [D\tau_i(T)]^{1/2} \ , \tag{3}$$

where

$$D = \tfrac{1}{2} v_F^2 \tau_0 = \tfrac{1}{2} v_F d \ . \tag{4}$$

Here D is the diffusion constant equal to 0.007 m^2/sec for a 100-Å Cu film.

We will show that the transition from 3D to 2D localization occurs at a temperature T_{cutoff} much higher than the temperature $T_{\min}$ at which the resistance is a minimum. We will further demonstrate that the minimum in the resistance $R(T)$ arises from two competing mechanisms—localization and inelastic electron-phonon scattering:

$$R(T) = R_0 + \Delta R_{\text{loc}}(T) + \Delta R_{\text{phonon}}(T) \ . \tag{5}$$

In the very thinnest films, we observed small deviations from the logarithmic temperature dependence of the resistance at lowest temperatures; the source of this deviation is assumed to come from spin-orbit scattering.

Sample preparation proved to be crucial since the observed localization effects were extremely sensitive to surface preparation and surface contamination. These observations are further supported by Bergmann's experimental investigations of impurities evaporated upon Mg films.[2] Copper was chosen since its films remain electrically conducting for thicknesses as small as 29 Å. Our Cu samples were prepared in an evaporator. The pressure was 10^{-5} Torr. The films were made by evaporating MRC 99.999% pure copper from a resistively heated tungsten boat onto standard glass microscope slides held at room temperature. The samples were 2 mm wide and 10 mm long.

Most of the samples in this paper were unprotected; they were measured immediately after preparation and then discarded. Some of the samples had a 500-Å SiO protective coating to reduce oxidation. With the SiO coating, the samples could be preserved for several months; the resistance typically increased by 10% each month. Extensive testing showed that the 500-Å SiO layer had no detrimental effect upon the localization properties. However, a thick 5000-Å protective layer of SiO greatly reduced the magnitude of the localization properties and decreased the resistance

of the sample; these effects possibly resulted from specular reflection of the electrons from the SiO layer. Moreover, any surface contamination (finger prints, frozen air, chemical reactants, thick layers of photoresist) dramatically reduced the localization effects.

Standard four-terminal dc measurements of the film were made using the Hewlett Packard 3456A digital voltmeter. The applied current was kept low enough so that the resistance was electric field independent (no heating effects); this resulted in a loss of resolution. However, greater resolution was achieved by using the statistical averaging functions of the Hewlett Packard (HP) instrument; 50 to 100 readings in the voltage ratio mode were made for each current direction to yield a single mean-resistance reading. Generally, the resistance could be resolved to within a few ppm.

A standard liquid-helium cryostat and calibrated Ge thermometer were employed for the measurements.

The elastic time τ_0 can be deduced from the residual conductance σ_0 in Eq. (2). The resistance varies as d^{-2} for a wide range of copper thicknesses between 70 Å $\leq d \leq$ 400 Å.[3] The $R \propto d^{-2}$ dependence strongly suggests that the elastic time τ_0 is thickness limited, $\tau_0 = d/v_F$ (Ref. 4); τ_0 takes on a typical small value of 7×10^{-25} sec in a 100-Å film. For film thicknesses less than 70 Å, the measured R increases more rapidly than the predicted d^{-2} value; this is presumably due to the inhomogeneous thickness of the films.[3] Two-dimensional localization should cease when the inelastic diffusion length L_i becomes equal to or less than the film thickness d; for smaller values of L_i 3D localization effects come into play. The cutoff temperature T_{cutoff} occurs when

$$L_i \simeq d \ . \tag{6}$$

Using the fact that $\tau_0 = d/v_F$ and Eq. (4), we find

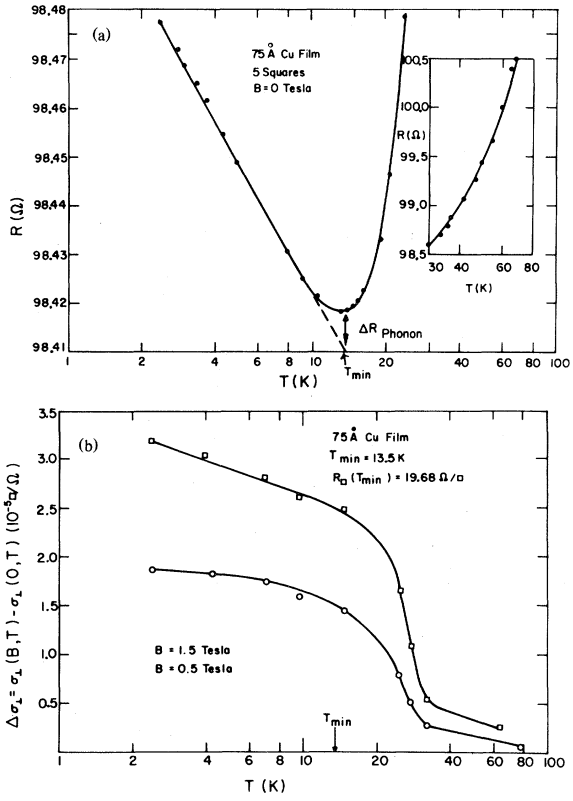

FIG. 1. The change of conductance $|\Delta\sigma| = \Delta R/R^2$ per temperature decade as a function of resistance. The mean value of αP is 1.7. In thick low-resistance films, magnetic impurities enhance $|\Delta\sigma|$.

that

$$\tau_i \simeq 2\tau_0 \ \text{ at } T_{\text{cutoff}} \ . \tag{7}$$

Thus the inelastic time must take on a value of

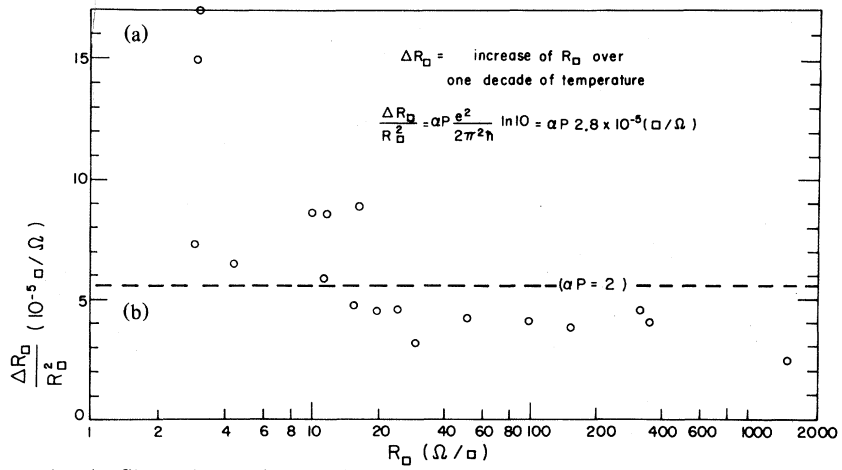

FIG. 2. (a) Upper graph—the film resistance is plotted against the logarithm of temperature. The change in conductance per temperature decade, given by Eq. (8), is equal to 4.6×10^{-5} □/Ω for this 75-Å-thick Cu film. The high-temperature phonon contribution to the resistance ΔR_{phonon} is also defined. T_{min} for this film is 13.5 K. (b) Lower graph—positive magnetoconductance as a function of the logarithm of temperature. Note that the negative MR continues to liquid-nitrogen temperatures even though $T_{\text{min}} = 13.5$ K.

$2\tau_0 \simeq 10^{-14}$ sec at T_{cutoff}. For our 100-Å films, we estimate this temperature to be about 150 K using $\tau_i = 3.4\times 10^{-10} T^{-2}$ obtained from our magnetoresistance data. The resistance minimum in a 100-Å film occurs at 11 K.

Experimentally, one can determine αP by measuring the logarithmic decrease of the sample conductance over one decade of temperature. Equation (1) predicts that

$$|\Delta\sigma| = \frac{\alpha P e^2}{2\pi^2 h} \ln 10 = \alpha P 2.83\times 10^{-5}\ \square/\Omega\ , \qquad (8)$$

where $\Delta\sigma$ is the decrease of conductance over a temperature decade. If the electron gas is noninteracting, then $\alpha = 1$. However, because of Coulomb interactions in metals, one would expect a value of α less than 1. Experimentally we observe from Fig. 1 that $|\Delta\sigma| = \Delta R/R^2 = 4.6\times 10^{-5}\ \square/\Omega$, thus suggesting that α takes on a value of 0.8 provided that $P = 2$. However, from these measurements it is impossible to determine P without knowing the value of α. As the sample thickness is increased above 150 Å, $\Delta\sigma$ increases to very large values (as large as $-45\times 10^{-5}\ \square/\ \Omega$). Magnetic impurities could cause this increase through the Kondo effect, which also yields a $R \propto \ln T$ dependence.[3]

It should also be possible to extract the exponent P from the high-temperature "phonon" resistance data [Eq. (5)] since

$$\Delta R_{\text{phonon}} \propto \tau_i^{-1} \langle 1-\cos\theta\rangle_{\text{av}}\ . \qquad (9)$$

The term $\langle 1-\cos\theta\rangle_{\text{av}}$ is the forward-scattering correction which accounts for the weak scattering of the electrons by the photons.[5] At low temperatures the forward-scattering correction goes as T^2. At high temperatures, the forward-scattering term approaches 1. For our films at low temperatures we would expect a temperature power fit of the form $\Delta R_{\text{phonon}} = aT^q$ to describe the data with $q = P+2$. At higher temperatures, we would intuitively expect a weaker temperature dependence with q taking on values considerably less than 4. Thus, if the phonon contribution to the film resistance can be extracted from the data, it should be possible to obtain a value for P. Writing explicitly Eq. (5) we get

$$R(T) = R_0 - R_0^2 \frac{\alpha P e^2}{2\pi^2 h} \ln T/T_{\text{cutoff}} + a(R_0) T^q\ . \qquad (10)$$

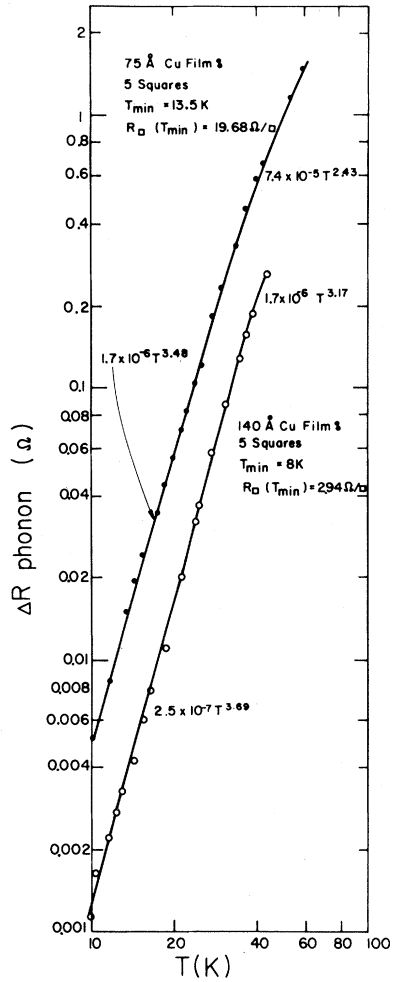

FIG. 3. Temperature dependence of $\Delta R_{\text{phonon}} = aT^q$ for two different Cu films. The exponent q takes on a typical value of 3.6 in the 10- to 20-K temperature interval. At higher temperatures q decreases to about 2.5.

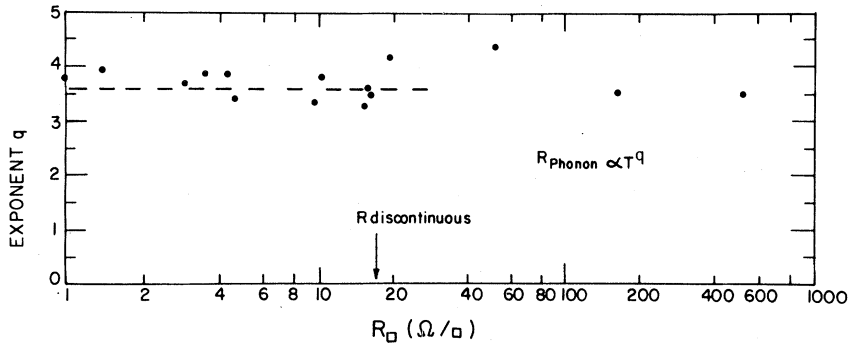

FIG. 4. Values of the exponent q obtained from films having different resistances R. For very low-resistance films, q tends to 4 suggesting that $P = 2$ for $\tau_i \propto T^{-P}$.

The second term extends to $T \leq T_{\text{cutoff}}$. Magnetoresistance measurements show that T_{cutoff} is much higher than T_{min}. The magnetoresistance remains negative even at liquid-nitrogen temperatures as shown in Fig. 2(b). Thus localization effects do extend to much higher temperatures than T_{min}, but in simple resistance measurements they are masked by the strong phonon contribution.

The unusual temperature dependence of the magnetoresistance (MR) data below 15 K arises from temperature-independent scattering due to spin-orbit interaction which dominates over the inelastic scattering; the MR data are treated in detail in another paper.[6]

The resistance data above T_{min} did not fit a power law in temperature unless the localization contribution was added.

Hence by extrapolating the logarithmic data to higher temperatures and subtracting off its value from the measured resistance [Fig. 2(a)], one obtains the ΔR_{phonon} data. Typical results are shown in Fig. 3 and clearly demonstrate the power-law dependence of the phonon contribution. This procedure was done on some 15 samples and the results appear in Fig. 4. Clearly for the low-resistance thick films, q tends to 4; these films have T_{min}'s of 5 to 7 K. We find that $p = q - 2 \simeq 1.9 \pm 0.3$.

The temperature T_{min} at which the resistance minimum occurs depends rather strongly upon the sample resistance as Fig. 5 indicates. By differentiating Eq. (10), one gets $R_0 \propto T_{\text{min}}^{q/2}$. From Fig. 5 we observe that $q \simeq 4$ and hence $P \simeq 2$.[7] For film resistances below $3\,\Omega/\square$, T_{min} is determined by the Kondo effect and no longer by localization. Moussouros and Kos found that 0.5-ppm Fe impurity in bulk Cu was sufficient to cause a resistance minimum at 4.5 K.[8]

Another anomalous effect was observed in the resistance behavior of the very thinnest films. These films showed T_{min}'s between 35 to 60 K and exhibited weaker logarithmic temperature dependence immediately below T_{min}. Typically, values of $|\Delta\sigma|$ per temperature decade were 3 to $4 \times 10^{-5}\ \square/\Omega$ as compared to values of 4.5 to $5.5 \times 10^{5}\ \square/\Omega$ for the thicker films. Moreover, as the temperature was decreased below 10 K, the experimental resistance fell

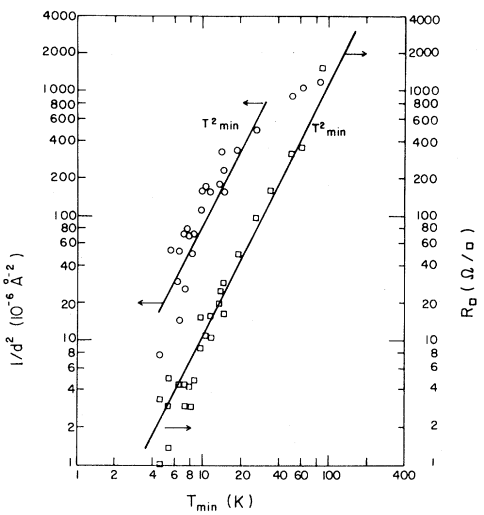

FIG. 5. Behavior of T_{min}, the temperature at which the resistance minimum occurs, for different film resistances R. The relation between T_{min} and d^{-2} is also shown where d is the film thickness. T_{min} saturates at 5 K owing to magnetic impurities which have a Kondo temperature at this temperature.

below the extrapolated logarithmic line. Maekawa and Fukuyama[9] have calculated the contribution of spin-orbit interaction to the conductance of the film. In zero magnetic field, another term is added to Eq. (1), σ_{so}, given by

$$\sigma_{\text{so}} = \frac{\alpha e^2}{2\pi^2 h} \frac{3}{2} \ln\left(1 + \frac{4\tau_i}{3\tau_{\text{so}}}\right), \tag{11}$$

where τ_{so} is the spin-orbit scattering time. From magnetoresistance data we obtain for the thinner films $\tau_{\text{so}} \simeq 2 \times 10^{-12}$ sec and $\tau_i(T) = 3.2 \times 10^{-10} T^{-2}$. At about 10 K τ_i becomes comparable to τ_{so} and spin-orbit interaction starts to destroy localization.

In conclusion, the zero-magnetic-field data for Cu films suggests that $P \simeq 2$, that 2D localization effects are present provided that $\tau_i > 2\tau_0$, and that spin-orbit scattering appears to cause deviations from the $\ln T$ dependence of the resistance. Unfortunately, we have not been able to extract the magnitude of τ_i from these data; it is best obtained from magnetoresistance measurements.

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[4]Values for the parameters are the following $m^* = 1.5 m_e$, $v_F = 1.4 \times 10^6$ m/sec, and $n = 8.5 \times 10^{28}/\text{m}^3$.

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