Brief Reports

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Localization in a three-dimensional metal

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The transition from a positive to a negative temperature coefficient of resistivity (TCR) has been studied in the artificially layered metallic system Nb-Cu. We find the TCR to undergo a rather sudden change in sign from positive to negative as the layer thickness is decreased below approximately 10 Å. The temperature dependence of the conductivity in the nonmetallic regime can be explained by localization-type theories, and implies that both localization and Coulomb effects are important in this purely metallic three-dimensional system.

The transition from a positive temperature coefficient of resistivity (TCR) ("metallic" behavior) to negative TCR ("nonmetallic") has received considerable attention in the last year.[1] This interest has been motivated by two classes of theories, localization[2] and electron-electron correlation[3] as well as by a large number of experimental papers. To date, this work has been predominantly in two-dimensional systems. Many of these experiments in "metals" have been performed in materials composed of mixtures of metals with insulators where, not surprisingly, as the insulator fraction is increased the electronic conduction progressively localizes. In contrast to this, very little attention has been given recently to experiments in three dimensions[4] or to systems of completely metallic components.[5]

We present data on an artificially layered metallic system: Nb and Cu. Since the constituents have a eutectic binary-phase diagram,[6] it is found that once the sample is prepared very little interdiffusion occurs. Above ~ 10-Å-layer thickness the material shows long-range structural coherence perpendicular to the film plane (z direction) and positive TCR. Below this thickness x-ray diffraction data show the superlattices to become progressively disordered and they undergo a rather sudden change to negative TCR. The details of this transition and the relation of the temperature dependence in the nonmetallic regime are the subject of the present Brief Report. We would like to stress that the present system has purely metallic constituents; therefore all the results are direct consequences of changes in the electronic mean-free path. This is in contrast to systems composed of mixtures of metallic and insulating components where changes are caused by the addition of progressively larger fractions of insulator into the metal. In spirit, the present experiments are similar to earlier resistivity studies in radiation-damaged materials.[7]

Nb-Cu superlattices were prepared using a sputtering technique described elsewhere.[8] Single-crystal (90° orientation) sapphire substrates were held on a rotating platform which moved them alternately between the two beams of sputtered Nb and Cu particles. In this manner samples of individual layer thickness in the range 3.6–5000 Å with total film thickness ~ 1 μm were prepared. As is discussed below, for layer thicknesses below 10 Å the material becomes progressively more disordered; however, in this regime the "layer thickness" remains a good parameter for characterizing the elastic mean-free path. This point is discussed in detail below. A detailed study of the x-ray structure shows that samples with layer thickness above 10 Å have long-range coherence (~ 10 superlattice-layer thicknesses) perpendicular to the layers (z direction) and ~ 100-Å crystallite size in the xy plane of the films. The x-ray-diffraction lines became progressively broader for samples below this thickness indicating that the material becomes more and more disordered. Above 10-Å-layer thickness, there is an inverse proportionality between the electrical resistivity and the layer thickness determined either from x-ray measurements or from the rotation speed of the platform along with the sputtering rate.[9] Thus, both methods may be used to determine the layer thickness in this thickness regime. Below 10 Å, where x rays can no longer be used to determine layer thickness, we will

use rotation speed as an indication of the mean-free path.

Figure 1 shows a graph of the low-temperature resistivity (20 K) versus layer thickness for a series of Nb-Cu samples. Above 10-Å-layer thickness the resistivity is inversely proportional to layer thickness as predicted for a mean-free path which is layer-thickness limited. Below this thickness the resistivity approaches saturation close to the Ioffe-Regel limit of 150 $\mu\Omega$ cm.

Figure 2 shows the temperature-dependent part of the resistivity for various layer thicknesses. Notice the systematic way in which the resistivity changes as a function of layer thickness. Again this shows that even in the region below 10 Å where the material is becoming disordered it is meaningful to use the rotation speed of the platform as an indication of the elastic mean-free path. In fact, the results in this Brief Report are solely a consequence of the controlled variation of the mean-free path in this artificially prepared metallic system. Layer thickness forms a useful parameter for characterizing the mean-free path, but the results are independent of the exact structure of our films. At any arbitrary fixed temperature anywhere in the range approximately $20 < T < 300$ K the TCR shows a rather sudden change from positive to negative value as a function of mean-free path as shown in Fig. 3. This change from positive to negative TCR occurs for a mean-free path which is changed by only 2–3 atomic spacings (~5–6 Å).

The temperature dependence in the nonmetallic regime has been the subject of extensive theoretical investigation.[2,3,10] It has been shown that due to interaction effects or localization the temperature-dependent part of the conductivity is given by

$$\frac{\delta\sigma}{\sigma} = 1 - \left(\frac{3}{2x}\right)\ln(1+x)\left(\frac{1}{N_0\pi^3}\frac{k_B}{(hD)^3}\right)^{1/2} T^{1/2}, \quad (1)$$

where D is the diffusion coefficient for electrons, N_0 is the density of states at the Fermi surface, and

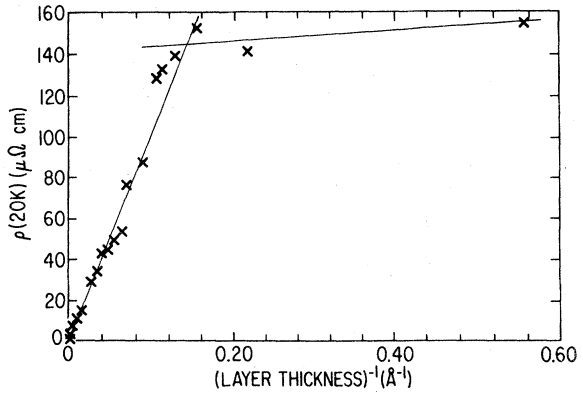

FIG. 1. Resistivity at 20 K vs inverse layer thickness.

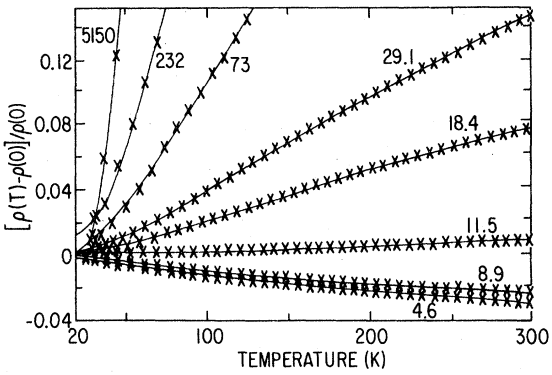

FIG. 2. Temperature-dependent part of the resistivity vs temperature for a number of samples of different layer thickness (labeled in Å on the curves). Zero-temperature values for resistivities $\rho(0)$ obtained from extrapolation of $\rho(T)$ vs T curves. Data from every second sample omitted for clarity.

$x = (2k_F/\mu)^2$, where k_F is the Fermi momentum and μ is the inverse screening length. Assuming $\delta\sigma \ll \sigma_0$, as we observe experimentally, and using the Einstein relation $\sigma_0 = N_0 e^2 D_0$ where σ_0 is the zero-temperature conductivity and D_0 is the zero-temperature diffusion coefficient, we obtain

$$\delta\sigma \cong 1 - \left(\frac{3}{2x}\right)\ln(1+x)\left(\frac{k_B e^4}{h^3\pi^6}\right)^{1/2}\frac{D_0}{D^{3/2}}T^{1/2}. \quad (2)$$

Figure 4 shows a graph of $\delta\sigma$ vs $T^{1/2}$ for a 9.5-Å sample. Above ~35 K the temperature dependence is well described by Eq. (2) with a temperature-independent diffusion coefficient. Similar behavior has been observed for all five other samples that show negative TCR. However, we would like to point out that in the same temperature region the dependence predicted by Mott's variable range-

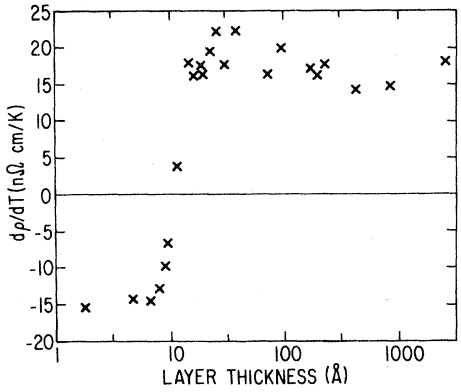

FIG. 3. $d\rho/dT$ at 150 K vs layer thickness. Similar curves are obtained for $d\rho/dT$ evaluated anywhere in the range $20 < T < 300$ K.

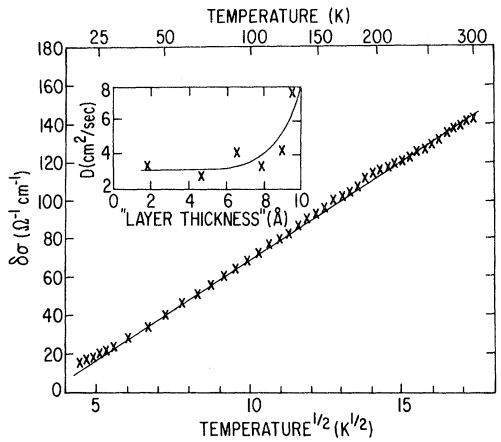

FIG. 4. $\delta\sigma$ vs $T^{1/2}$ for the 9.5-Å sample. Here $\delta\sigma = \sigma(T) - 7762.3\ \Omega^{-1}\text{cm}^{-1}$ is the change in conductivity from its value at zero temperature [obtained from extrapolating its $\rho(T)$ vs T curve]. Inset shows the layer-thickness dependence of the high-temperature diffusion coefficient extracted from Eq. (1).

hopping model[11] cannot be distinguished from the $T^{1/2}$ dependence, and therefore it also fits the experimental data. No other theoretical expression has been found to fit the experimental results.

The prefactor to $T^{1/2}$ in Eq. (1) can be extracted from a plot similar to Fig. (4). A "low-temperature" diffusion coefficient D_0 can also be obtained from the Einstein relation and the extrapolated zero-temperature conductivity σ_0. A comparison of these two quantities indicates that $[1-(3/2x)\ln(1+x)] \sim 10^{-2}$. Solving for x we find $k_F/\mu \simeq \frac{1}{2}$. This implies that both localization and correlation effects are important in the Nb-Cu system.[10] It is interesting to note that a plot of the "high-temperature" diffusion coefficient as extracted from the slope of Eq. (1) versus layer thickness (inset Fig. 4) shows the high-temperature diffusion coefficient to increase markedly as the nonmetallic-to-metallic transition is approached. This might be the origin for phonon softening[12] observed earlier in Brillouin scattering measurements.[13]

At this point we should mention that $T^{1/2}$ dependences have been observed earlier in granular aluminum which, in contrast to our system, is a mixture of insulator (Al_2O_3) and metal (Al).[14] We have reexamined the published[15] conductivities of radiation-damaged $LuRh_4B_4$, and we find it to show similar behavior in the same temperature range.

In summary, we have measured the temperature dependence of the resistivity for a series of three-dimensional short-mean-free-path metals. We find a rather sharp transition from metallic- to nonmetallic-type behavior as the mean-free path is reduced. The temperature dependence of the conductivity can be explained using localization-type theories and implies that both localization and Coulomb effects are important. We would like to emphasize that the present work involves a purely metallic three-dimensional system and therefore all effects are due only to changes in the mean-free path.

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