Localization and Size Effects in Single-Crystal Au Films

P. Chaudhari, H.-U. Habermeier,[a] and S. Maekawa[b]
IBM T. J. Watson Research Center, Yorktown Heights, New York 10598
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We show that localization effects can be significantly enhanced in the presence of quantum size effects. We have made measurements on single-crystal gold films of thicknesses less than 10 nm. An enhanced logarithmic temperature dependence of the resistance and a negative magnetoresistance provide qualitative support for this model.

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A number of experimental data on the transport properties of wires or thin metallic films at low temperatures have recently been interpreted in terms of localization and Coulomb interaction theories.[1] In the transport behavior of samples with restricted geometries, there is, however, another effect that can give rise to anomalous behavior. This is due to size effects—both classical and quantum. These are particularly important when the elastic mean free path of the electrons exceeds the geometrical size of the sample. This has been studied for a number of decades.[2] In this note we show that the combination of size effects and localization can lead to unexpected behavior which neither of the two separately predicts. Qualitative support for the ideas presented here is provided by the study of single-crystal gold films.

The logarithmic temperature dependence of the resistance and the magnetoresistance of thin films are believed to be well understood as resulting from localization and/or the interaction effect. The conductance is fitted by the equation[3]

$$\sigma' = (-e^2/2\pi^2\hbar)\alpha_T \ln T, \tag{1}$$

where T is the (normalized) temperature and α_T is a measure of the microscopic properties of the metal. Localization[3,4] and interaction theories[5,6] both predict a value of α_T close to 1. If spin-orbit scattering is strong, localization theories predict a negative value of α_T. Additional information about the mechanism controlling transport can be obtained from magnetoresistance. In a magnetic field perpendicular to the film, quantum intereference due to localization is suppressed, which leads to negative magnetoresistance.[7] In the presence of strong spin-orbit coupling, however, magnetoresistance is positive.[8] In contrast to localization, the interaction effect is not sensitive to weak magnetic fields.[4-6]

As the geometrical size of the wires and films is small, it is possible that the elastic mean free path of electrons can be comparable to or larger than the dimension of the conductor and the effect of surfaces should become increasingly important. Consider, for example, the case when there is complete specular reflection from a two-dimensional film. A free-electron model gives the following eigenfunction, which is labeled by the quantum numbers k_x, k_y, and $k = \pi n/d$, where d is the film thickness and $n = 1, 2, 3, \ldots$:

$$\Psi_{k_x,k_y,k} = \Phi_k(z)\exp[i(k_x x + k_y y)],$$
$$\Phi_k(z) = \sin(kz). \tag{2}$$

In a film with $d = 10$ nm, this leads to an opening in the conduction band which is estimated to be $\hbar^2\pi^2/2md^2k_B = 43.6$ K. In real metal films, electron correlation and exchange interactions can modify states near the surface.[9] For example, according to a self-consistent calculation of the energy bands in Cu films with (001) surfaces carried out for up to nineteen atomic layers,[10] the electronic states at the Fermi level (ϵ_F) were found to be built up of s and p electrons whereas the $3d$ electrons exist below ϵ_F. The number of conduction bands at ϵ_F was proportional to the number of atomic layers in the film. The free-electron model is therefore useful only for a semiquantitative discussion.

When electrons are specularly reflected from surfaces during the time interval between inelastic events, the quantum size effect (QSE) contributes to the anomalous conductance associated with localization. Its magnitude is proportional to the number of channels for electron diffusion. The anomalous conductance, expressed as the sum of the contributions from each band, is given by

$$\sigma' = (e^2/2\pi^2\hbar)n_0 p \ln T, \tag{3}$$

where n_0 is the number of the bands at the Fermi surface and the inelastic-scattering time is expressed as $\tau_\epsilon \propto T^{-p}$, with p being a constant. Since n_0 can be large, the $\ln T$ term in the conductance can be strongly enhanced by the QSE. In real thin films, interband scattering will tend to smear out the quantized nature of the band structure, which results in a reduction of the prefactor of the logarithmic term in σ'. For example, in the case of Si–metal-oxide-semiconductor structures, intervalley scattering partially washes out the effect of valley degeneracy.[11] In metallic films, as the intensity of interband scattering increases, we expect the number of channels to decrease to a point where only a single effective channel is observed. Thus, quantum size effects will be important only in relatively clean systems. We also anticipate spin-orbit scattering to be minimized in clean systems both because the density of defects associated with atomic scale structure in single-crystal films is lower than in the usually studied gold films and also because of specular reflection associated with a smooth surface. If reflection from surfaces is not completely specular but contains a diffuse component, then both the classical size effect and spin-orbit scattering are expected to be present. In particular, there can be a large magnetoresistance contribution from classical size effects.[2] It is also of interest to note that the QSE plays no significant role in the interaction term.

In order to search for these effects, we have prepared single-crystal gold films with smooth surfaces so as to maximize the possibility of specular reflection. Also, Au has been shown by a number of investigators to have a large spin-orbit scattering contribution.[12] If our ideas apply to the single-crystal Au films, there should be no significant spin-orbit scattering observed. This would manifest itself as a negative magnetoresistance in a perpendicular magnetic field.

Gold films less than 10 nm in thickness were grown by electron-beam evaporation in a high-vacuum system with a base pressure of 10^{-5} Pa during evaporation. To prepare smooth self-supporting single-crystal gold fims, a 1-μm-thick silver film was evaporated first onto a cleaved NaCl single crystal held at 300 °C during evaporation. The silver forms a continuous single-crystal layer, and its thickness smooths out the cleavage steps of the NaCl substrate. To reduce interdiffusion effects, the gold film was evaporated after lowering of the substrate temperature to 150 °C. The gold deposits epitaxially on the silver. The film quality was checked after evaporation by Laue back-reflection techniques and a study of selected area channeling patterns (Kikuchi patterns) in a scanning electron microscope. After this, the substrates were cut into slabs approximately 1×4 mm^2. The NaCl was dissolved in deionized water and the Ag film in dilute HNO_3. The remaining self-supporting gold film was washed in deionized water again and picked up either on a sapphire single crystal for electrical measurements or on a copper grid for transmission electron microscopy investigations. The transmission electron microscopy results confirmed that the films were smooth and single crystal. The films showed the presence of occasional twins and dislocations whose concentration varied from sample to sample. The precise thickness of the final Au film could not be determined but is believed to be less than 10 nm from rate monitors used during evaporation.[13]

In order to make a four-probe resistance measurement, silver epoxy was employed as a contact material. Finally, the specimens were inspected in an optical microscope with regard to shape and the presence of

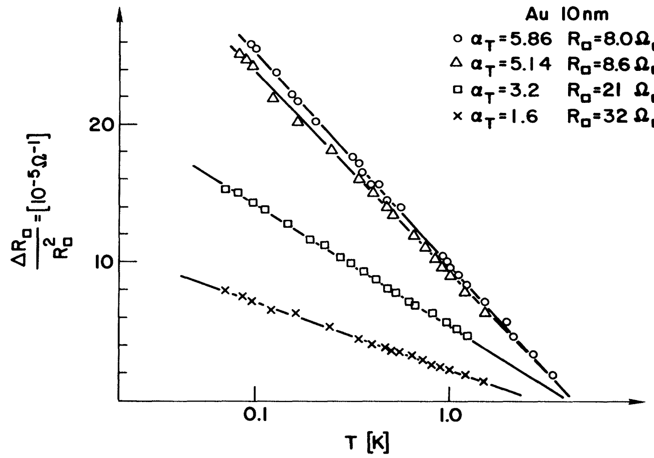

FIG. 1. Temperature dependence of the sheet resistance of single-crystal gold films for different values of the sheet resistance.

cracks. The specimens were mounted onto the cooling stage of a Leybold dilution refrigerator equipped with a superconducting solenoid. Measurements were carried out with a low-frequency ac lock-in technique bridge with probe currents less than 1 μA. The electronics was set up in a screened room, and changes in resistance of 1 part in 10^5 could be readily detected. The experimental results for the temperature dependence of the sheet resistance is given in Fig. 1. All samples investigated showed a negative slope for the ΔR vs $\ln T$ plot in the temperature range 40 mK $< T <$ 4.4 K. The slope depends strongly on the actual sheet resistance—the lower the sheet resistance at 4.4 K, the larger the slope. Typical magnetoresistance data are given in Fig. 2 for a temperature of 103 mK. The sheet resistance in parallel and perpendicular fields is anisotropic. In parallel fields (current parallel to the magnetic field) we always observe a positive contribution to the magnetoresistance. The transverse magnetoresistance (magnetic field perpendicular to the plane of the film) shows a negative value for low-sheet-resistance samples in the temperature range $T <$ 200 mK and magnetic fields $H <$ 0.8 T. With increasing sheet resistance and increasing temperature, the negative contributions to the magnetoresistance are reduced and vanish for $R_□ > 20\ \Omega_□$, even at the lowest temperature attainable in the dilution refrigerator that we used (40 mK). The positive magnetoresistance for larger magnetic fields ($H >$ 1 T) was found to be a function of the orientation of the sample with respect to the magnetic field and varied only slightly with temperature.[14] We observed an increase of about 4% from 0.095 to 7 K.

Values of α_T larger than those expected from localization and interaction effects have been reported by a

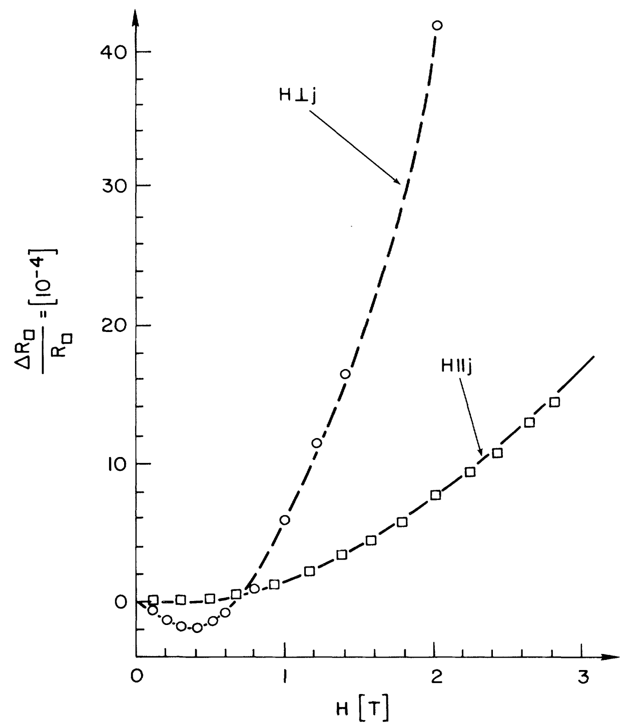

FIG. 2. Magnetoresistance of a film of relatively low sheet resistance (8 $\Omega_\square$). In the perpendicular case the magnetic field is perpendicular to the plane of the film.

number of authors.[15-17] These have been attributed to the Kondo effect. In our samples the magnetoresistance is anisotropic and, in the case of perpendicular field, negative. These results rule out the possibility of a significant contribution to the resistivity from the Kondo effect. The presence of a negative magnetoresistance and its temperature dependence is consistent with localization theories and is evidence that spin-orbit scattering is small relative to inelastic mechanisms.

In summary, we stress that the transport behavior of samples with restricted geometries is influenced not only by localization and Coulomb effects but also by size effects, both classical and quantum.[18] In the clean limit, size effects can dominate. We believe that our single-crystal Au films provide the first example of this effect.

[(a)]Permanent address: Max-Planck-Institut für Festkörperforschung, Stuttgart, Federal Republic of Germany.

[(b)]Permanent address: The Research Institute for Iron, Steel and Other Metals, Tohoku University, Sendai 980, Japan.

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[13]We are reluctant to claim a precise thickness because of concern that there might have been interdiffusion between Au and Ag. The alloy would be removed by the nitric acid leaving us with a smaller amount of Au than the nominal thickness indicated by the rate monitor.

[14]The perpendicular magnetoreistance data could be fitted approximately by the classical-size-effect equation and yielded a mean free path of elastic scattering of approximately 700 nm. This proved to be useful as it provided us with some confidence that we were indeed in the regime where the elastic mean free path is greater than the film thickness. After subtraction of the classical-size-effect contributions to the magnetoresistance, the remaining magnetoresistance data could be fitted by the two-dimensional localization formulas provided that the value of n_0 deduced from the temperature dependence of the resistance was used.

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[18]Since the QSE results in two-dimensional electronic states, it may induce the quantized Hall effect in a higher magnetic field. Many degenerate bands will contribute to the quantized Hall effect independently so that a kind of reproducible noise in the magnetoresistance may be expected.