Direct Measurement of Potential Steps at Grain Boundaries in the Presence of Current Flow

J. R. Kirtley, S. Washburn, and M. J. Brady
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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We have used a new technique to measure simultaneously the surface topography and surface potential of current-carrying polycrystalline $Au_{60}Pd_{40}$ thin films using a scanning tunneling microscope. The variations of the gradients of the surface potential from a macroscopically constant value which are associated with scattering from grain boundaries in these films are observed. We find that the local potential changes abruptly at the boundaries between the grains.

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The residual resistivity of metals results from collisions between the current carriers and deviations from periodicity in the lattice potential. When carriers impinge on a potential barrier, charge accumulates on the near side of the barrier and depletes on the far side. The "dipole" field which results is the ultimate source of resistance in disordered material.[1] The gradient in the chemical potential of the carriers along a current-carrying wire, while macroscopically constant, comprises microscopic variations that reflect the positions of the scattering sites (such as impurity atoms, vacancies, dislocations, etc.) which resist the flow of carriers. The voltage is "dropped" in the collisions with impurities. This picture is in contrast to the conventional Boltzmann picture of transport, in which the details of the impurity potential are ignored completely or removed from the problem by averages, and consequently the potential is taken to have a constant gradient along the sample. The picture of localized changes in the carrier potential has, however, gained wide acceptance among theoreticians.[2] The theoretical description of quantum-mechanical effects in the conductivity of metals has relied on models which incorporate the local fluctuations in the chemical potential which result from impurity scattering.[3,4] To date no experimental observations of such local fluctuations have been made.

In this Letter, we describe measurements of the surface potential of a polycrystalline metal film (60-nm-thick $Au_{60}Pd_{40}$) which reveal essentially steplike changes near the boundaries between the grains. When current is flowing through the film, the potential exhibits terraces which are spatially correlated with the metal grains observed in simultaneous measurements of the film's topography. In the absence of current flow, no such terraces are observed, and the surface potential is essentially flat. We therefore associate the steplike drops in the surface potential with the changes in the local chemical potential as the carriers move along the sample—they are the fundamental source of the resistance $\Delta V/I$ of the sample. For these experiments, we used a method somewhat different from that of the pioneering experiments of Muralt and co-workers, who observed the potential gradients in discontinuous gold films[5] and in the vicinity of hetero-junction barriers[6] by using a scanning tunneling microscope[7] as a potentiometer.

The samples were mounted in a single-piezo-tube, differential spring-approach, scanning tunneling microscope, which has been described in detail elsewhere.[8] All of the scanning-tunneling-microscope measurements reported here were made at room temperature. The sample chamber was pumped out to $\simeq 10^{-6}$ Torr with a turbomolecular pump, sealed off, and then disconnected from the pump to minimize vibrations during the measurements. The pressure in the chamber stayed below 10^{-4} Torr at all times. The measurements were made on 60% Au, 40% Pd ($Au_{60}Pd_{40}$) films sputtered onto oxidized silicon wafers through metal shadow masks. Two silver-paint contacts defined the length of the film, which was 450 μm long, 220 μm wide, and 600 Å thick. The films had rather high resistances of about 350 $\Omega/\square$, and resistivities of about 2000 $\mu\Omega$-cm. This is about an order of magnitude larger than typical resistivities for disordered films[9] and about 2 orders of magnitude larger than the bulk resistivity of the alloy. The reason for this high resistivity is not understood. In spite of the large values of resistivity, the films are good metals—the film resistance is far below the metal-insulator transition value, and the resistance is nearly independent of temperature (decreasing by a few percent by $T=5$ K) and independent of the current through the film. Estimates of mean free paths from the resistivities of these films yield unrealistically short values (shorter than the lattice spacings). In contrast, estimates for the reflectivity of the grain boundaries from Landauer's formalism,[1] given the measured film resistivities, yield values close to 1. The self-consistent picture can then be drawn in which the resistance of the films is dominated by grain boundaries which scatter electrons strongly. This picture is supported by our potentiometric measurements.

Our method used for the measurement of the local surface potential takes advantage of the interrupted feedback technique of Feenstra and co-workers.[8,10] In this method (see Fig. 1) the tunneling tip is connected to virtual ground through a current-sensitive preamplifier. The sample is biased with a square-wave voltage train. The potentiometer is adjusted so that the tip-sample po-

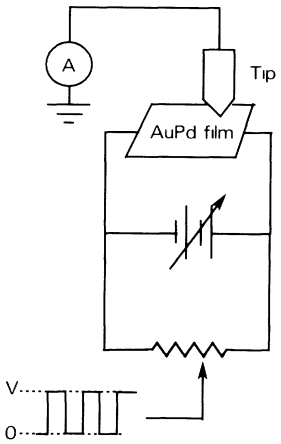

FIG. 1. Schematic of sample-biasing technique used in these measurements.

tential is zero when the center tap is at ground potential. This allows us to adjust the local potential between the tunneling tip and the sample to within a few tens of microvolts while applying several volts between the two ends of the film. In the portion of the cycle when the sample is biased at a voltage V, the feedback loop to the z piezo is held closed, and the surface topography is measured in the standard way.[7] The feedback loop is then interrupted, the sample bias is brought to zero, and the tunneling current is measured. The tip is then moved to a different point on the film, and the cycle is repeated to measure simultaneously the surface topography and potential.

Since we are tunneling into a metal at small potential differences, the tunneling current-voltage characteristic is linear, and V_S, the local potential of the sample relative to the tip, is given by

$$V_S = -I_0 R = -I_0 V/(I_1 - I_0). \quad (1)$$

I_0 is the current at zero sample bias, I_1 is the current at sample bias V, V is the amplitude of the biasing square wave, and R is the tunneling resistance.

Our technique has two advantages. First, it is simpler to implement than the double feedback technique of Muralt and Pohl.[5] Second, signal-to-noise ratios at least 10 times better are attained, because the dominant source of noise in these measurements is fluctuations in the tunneling resistance. Therefore the noise in the measured potential is minimized when the potential measurements are made with zero voltage between the tip and sample. Muralt and Pohl[5] used a modulation voltage centered at zero bias, but needed appreciable modulation amplitudes. Our square-wave bias allows topography to be recorded at finite sample voltage (under relatively high noise conditions) while the potential is measured at low voltage (and low noise). This technique has the disadvantage that the topography is not measured at constant tunneling resistance, because, although the tunneling

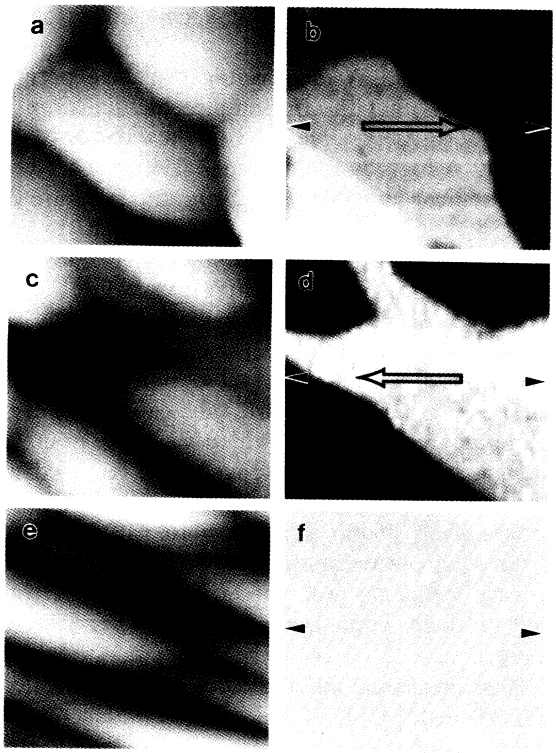

FIG. 2. Topographic (left column) and potentiometric (right column) images of $Au_{60}Pd_{40}$ film under fields of 85, -85, and 2 V/cm. Each pair of topographic and potentiometric images were taken simultaneously. Because of thermal and piezo drifts, each pair of images was of a different region of the film. The topographic images are grey-scale images with 237, 90, and 184 Å from white (high) to black (low) for (a), (c), and (e), respectively. The potentiometric images [(b), (d), and (f)] are on a grey scale of 450 μV from white (positive) to black (negative).

current at sample bias V is held constant, local variations in the surface potential cause the tip-sample potential to change, changing the tunneling resistance. However, since the tunneling resistance depends exponentially on the tip-sample distance, the distortions of the topographical images due to the variation in potential are expected to be very small. The variations in height in our measurements were of order 100 Å; the largest distortions of the images that could be expected from the variations in tunneling resistance were $\lesssim 10$ Å.

Figure 2 shows topographic (left column) and potentiometric (right column) images acquired simultaneously with the technique described above for one of our films. In Figs. 2(a) and 2(b) a transverse electric field along the film of $E_f = 85$ V/cm was applied, with the scan and current directions oriented in such a way that the average voltage [indicated by the arrow on 2(b)] was from left to right. The data of Figs. 2(c) and 2(d) were taken with $E_f = -85$ V/cm, with the average voltage drop from right to left. Figures 2(e) and 2(f) were taken with

much smaller average field of 2 V/cm across the film from left to right. This small field of 2 V/cm was used to compensate for small residual voltages in the experimental apparatus, so that the tip-sample potential drop was as close to zero as possible. All of the images covered areas of 240 Å high by 250 Å wide. The grey scales for the topographical images [2(a), 2(c), and 2(e)] are 237, 90, and 184 Å, respectively, from white (high) to black (low). The scales for the potentiometric images [2(b), 2(d), and 2(f)] are 450 μV from positive (white) to negative (black) of the sample with respect to the tip. The last two images, 2(e) and 2(f), were taken immediately after the z-piezo drive was retracted and extended, and residual creep in the piezoelectric tube caused the distortion of the topographic image which made the grains appear elongated. All of these images were recorded with a square-wave amplitude of $V=1$ mV. Similar results, with progressively poorer signal-to-noise ratios, were obtained for square-wave amplitudes up to 20 mV. The square-wave frequency was chosen to be 20 Hz, corresponding to about 8-min acquisition time per 100×100 pixel image.

For these measurements the current was controlled at $I_1=100$ pA in the half of the cycle in which the feedback loop was on, and the grains in these films were 50 to 100 Å in diameter, so that the density of current tunneling into a particular grain was about 100 A/cm^2. In contrast, the externally applied current density in the films was of order 3×10^7 A/cm^2. Given this disparity of 5 orders of magnitude between the applied current and the tunneling current, we do not believe that the tunneling current contributed an appreciable perturbation of the potential distribution set up by the transverse film current. If certain grains were nearly isolated from the rest of the film (so that the resistance between the particular grain and the rest of the film was larger than the tunneling resistance), then the discharging of the probe current from these grains would be poor. In this case, the extra accumulated charge *would* yield significant perturbations in the measured surface potential. However, since we operated at tunneling resistances of at least 10^7 Ω, while the film resistances were about 350 Ω/□, effects due to charging of individual grains seem unlikely. Even the small remnant perturbation from the electrostatic potential between the tip and the sample current should have been irrelevant in the sense that it was constant as long as the probe tip did not change shape, and simply tracked along with the tip to add a net constant offset to the measured potential.

The images of Fig. 2 make it clear that when there is current running through the film [Figs. 2(b) and 2(d)] there are well-defined plateaus in V_S that correspond to individual grains in the topographic images. When much smaller currents flow through the film [Fig. 2(f)] there is essentially no variation in the measured surface potential. The spatial correlation of the terraces in the surface potential with the grains in the topographical image, and the absence of such terraces when no current flowed through the film, lead us to the conclusion that V_S is, in fact, a measure of the local chemical potential of the carriers.

We show in Fig. 3 cross sections through the potential images of Figs. 2(b), 2(d), and 2(f) along the cuts marked by the arrow heads. Those cross sections are labeled in the figure by the average fields across the films ($E_f=85$, -85, and 2 V/cm, respectively). Also included in this figure are dashed curves illustrating the relative potential variations that would be expected from the average field across the sample. The zero in potential in each of the experimental curves was set by the potentiometer. The curve corresponding to $E_f=2$ V/cm has been offset by -50 μV for clarity. The zeros in potential for the dashed curves were chosen arbitrarily. It can be seen clearly from these figures that the bulk of the voltage drops occur at spatially well-defined points (corresponding to the grain boundaries in the topographical images of Fig. 2). The total potential drops across the images have the correct sign and approximately the correct amplitude expected from the average electric fields across the films. Transport through these films, however, appears to be highly spatially inhomogeneous. For example, the potential drop in the upper portion of Fig. 2(d) is not monotonic, indicating that there are large inhomogeneities in the transmission coefficients between the grain boundaries. The local electric fields are about an order of magnitude smaller inside the grains than at the grain boundaries. The small gradients within the grains probably result from phonon scattering. This supposition is consistent with the 10% drop in resistance observed as the temperature was reduced from 300 to 5 K.

As expected from the analysis above, the noise in the

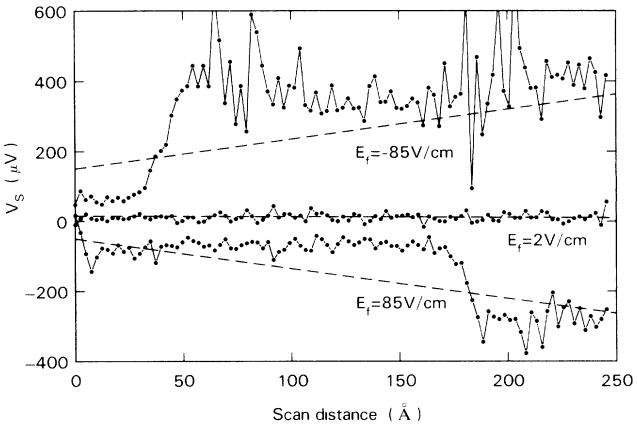

FIG. 3. Cross-sectional cuts along the center lines (marked by the arrow heads) of the potentiometric images Figs. 2(b), 2(d), and 2(f), corresponding to transverse film fields E_f of 85, -85, and 2 V/cm, respectively. The dashed lines represent the expected potential drops from the average fields across the sample.

potential measurements becomes larger the farther the sample potential is from zero. If we define the noise in the potential signal to be the average of the absolute values of the differences in measured potential from point to point in a particular scan, the noise level in Fig. 2(f) (E_f=2 V/cm) is 11 μV. It is the unprecedented noise level allowed by our new biasing technique that makes it possible to observe these small potential steps.

The steps in surface potential between grains are sharp; the change from plateau to plateau occurs over a length scale of $\lesssim$10 Å. Between these steps the potential plateaus are nearly flat. The expected rounding of the potential steps from the space charge near the barriers[11,1] should decay on lengths of the order of the screening length. We estimate that the screening length for our film is $l_s \simeq 4$ Å, which is near the lateral resolution of the microscope. There *is* some rounding of the steps in Fig. 3 on length scales of about 5 Å, but given the noise level and the rough lateral resolution, we cannot conclude that these are the signatures of space charge near the grain boundaries.

The traces in Fig. 3 bear a strong resemblance to predictions for transport through a one-dimensional disordered medium[12] in the case where the scattering events are independent. (See, for instance, Fig. 9 of Ref. 11.) The assumption that the scattering events are independent is appropriate for high temperatures where the phase memory length L_ϕ of the carriers is short. At lower temperatures, when L_ϕ grows to be larger than the average grain size, the scattering events will no longer be independent. Interference among the carrier wave functions will lead to correlation in their motion, and according to recent theory,[4] the correlations in carrier motion have a length scale L_ϕ. We expect that this correlation will change the surface potential profiles in our films, and experiments to study this temperature dependence are currently under way.

In conclusion, we have used a new technique to measure the current-induced potential variations in thin polycrystalline metal films. These measurements indicate that a large fraction of the total potential drops occur at the grain boundaries. This result is consistent with theoretical work on the role of impurity and boundary scattering on transport in metals.

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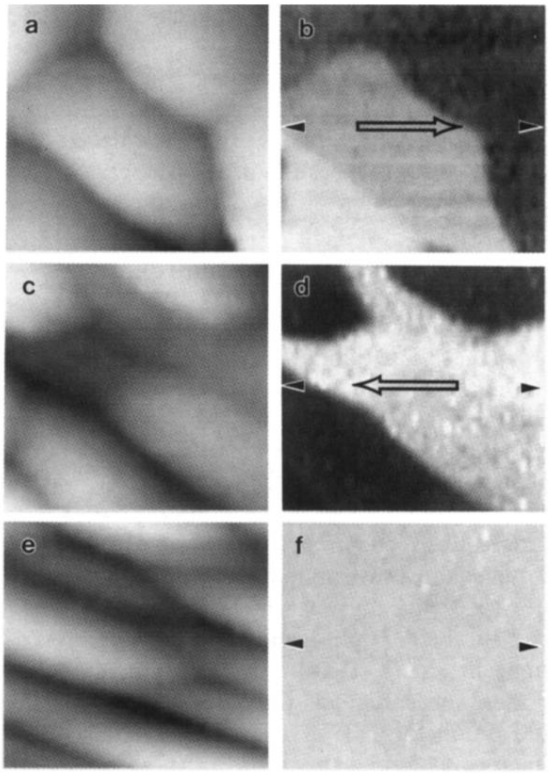

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