Electrical and mechanical properties of metallic nanowires: Conductance quantization and localization

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Measurements of room-temperature electronic transport in pulled metallic nanowires are presented, demonstrating that the conductance characteristics depend on the length, lateral dimensions, state and degree of disorder, and elongation mechanism of the wire. Conductance during elongation of short wires, $l \sim 50$ Å, exhibits periodic quantization steps with characteristic dips, correlating with the order-disorder states of layers of atoms in the wire, predicted via molecular dynamics simulations. The resistance of longer wires, $l \gtrsim 100$ Å, exhibits localization characteristics with $\ln R(l) \sim l^2$. Effects of disorder and variations in wire geometry, exhibited via their influence on the transmittivity of the conductance channels and/or the quantization conditions, are demonstrated. © *1995 American Vacuum Society.*

I. INTRODUCTION

Systems of small size or low dimensionality may, and often do, exhibit properties different from those found in the bulk, the investigation of which may open avenues for discovery of new physical phenomena. The electrical conductance was one of the properties that showed new behavior to be measured in such systems. It was found that the conductance of a small constriction in a two-dimensional electron gas (2DEG) under cryogenic conditions is quantized in units of $2e^2/h$ when the constriction diameter is varied.[1,2] This phenomenon is not restricted to a 2DEG but should also occur in three-dimensional (3D) metallic point contacts with small constriction diameters.[3] Another property of 3D nanowires pertaining to their mechanical behavior was predicted via molecular dynamics (MD) simulations.[4] In these simulations it was observed that elongation of solid gold junctions between a tip and a sample consists of a sequence of stepwise layer-by-layer stages, which were predicted to result in oscillatory behavior of the recorded force, with approximate interlayer distance periodicity.

Recently, the scanning tunneling microscope (STM) has been used to fabricate structures of atomic dimensions. By using an STM with a gold tip and sample, point contact measurements at ambient pressure and room temperature showed jumps in the current when the contact was elongated.[5] The conductance steps occurred in multiples of $2e^2/h$ and are attributed to quantization of the conductance. Similar results have been reported using an STM in ultrahigh vacuum (UHV) with Ni, Cu, and Pt samples,[6(a)] and with Pb,[6(b)] as well as in measurements employing a mechanically controllable break junction technique.[7]

In this paper we describe studies pertaining to the fabrication and properties of metallic nanowires of variable thicknesses and lengths. Preparation of such wires was achieved by controlling the elongation process carefully using digital feedback. By reducing the retraction speed of the tip when stretching the contact, we have been able to fabricate longer wires. In these long wires ballistic transport may disappear, and diffusive transport due to increased disordering should be more appropriate for a description of the conductance properties. We find[8] that the resistance of long ($l \gtrsim 100$ Å) and narrow wires exhibits localization behavior with an exponential dependence of the resistance (R) on the square of the wire length (l), i.e., $\ln R \sim l^2$.

Moreover, the combination of electronic conductance measurements with MD simulations of the wire elongation processes[4] allows us to elucidate the physical origins of the observed patterns.[8] For short wires ($l \approx 50$ Å), the periodic occurrence of quantized conductance accompanied by characteristic ''dips'' (local conductance minima associated with the presence of disorder), are correlated with atomic-scale structural transformations occurring during the layer-by-layer order–disorder elongation process.[4]

II. EXPERIMENTAL

Experiments were performed at room temperature using a homemade STM head, operating under ambient conditions (similar results were obtained in UHV). Gold evaporated onto mica was used as a sample, with either Pt/Ir or gold tips (the results are insensitive to the kind of tip used; most likely, the Pt/Ir tip apex is covered by gold atoms once the tip touches the sample.)[4] In order to achieve optimal control on the STM operations, we have developed a digital control unit. In this way, we can break the feedback control loop at any desired time and act on the piezos, as well as vary other system parameters.

Electrical contact is produced starting from normal tunneling conditions by applying a short-voltage pulse of a few microseconds as explained in Ref. 5 or by indentation. The feedback loop is open just before the pulse occurs, and the tip is slowly withdrawn, by acting on the z-piezo voltage, at rates of a few angstroms per second. The current flowing through the contact is measured by means of a double I/V converter connected to a high quality analog switch described elsewhere.[5]

III. RESULTS AND DISCUSSION

We show first data corresponding to long wires. In our experiments a gold tip was brought into contact with a gold sample starting from tunneling conditions. Subsequently the contact was elongated by withdrawing the z-piezo to an initial distance of 95 Å. A constant bias voltage of 500 mV was applied during this process. The contact resistance at this point was already 15 kΩ. In Fig. 1 we display the behavior of the resistance and current as the wire is elongated an additional 20 Å. The resistance values (from 15 to 37 kΩ) are considerably higher than 12.9 kΩ, the resistance of one quantum conductance channel.

In other experiments, values higher than 100 kΩ were observed, with the sample and the tip still in mechanical contact. In this case, a Pt/Ir tip was retraced for a total distance of 185 Å until a break occurred. The bias voltage applied was 100 mV, and the resistance reached a value of 150 kΩ. After the breaking process, we measured the distance traveled by the tip until the onset of the tunnel current, obtaining a value of 200 Å for the z displacement.

In such nanowires Anderson localization,[9] i.e., $R(l)\sim\exp(l/L)$, may occur when the length of the wire l exceeds the localization length L, and is smaller than the phase-breaking length L_ϕ (and in any case $L_\phi > L$), with the localization length given by[10] $L\sim Sk_F^2\lambda$, where S is the cross-sectional area of the wire, k_F the Fermi wave vector, and λ the elastic mean-free path. Note that in very narrow wires, as in our case, the number of transverse modes (channels) is small and thus the localization length may be of the order of λ. Under such conditions localization in systems characterized by small values of the elastic mean-free path may be expected even at room temperature. As the total volume Ω of the connecting wire remains constant during the elongation process (as observed in MD simulations)[4] S will decrease with increasing l (e.g., for a cylindrical wire $S=\Omega/l$). This leads to the $\ln R\sim l^2$ dependence observed in Fig. 1, which serves as an a posteriori confirmation of our interpretation of the data in the localization framework.[8]

For shorter narrow wires (i.e., $l<L$) the conditions for quantization may be reached,[3,5,11–13] as shown in Fig. 2. In this case a Pt/Ir tip was brought into contact with a gold surface; the applied bias voltage was 32 mV. We observe quantized steps in the conductance (in one or two units of $2e^2/h$), separated by length intervals (or combinations thereof) of approximately 2 Å, and the occurrence of conductance minima (dips) which tend to accompany the quantized jumps in the conductance.

To discuss the above patterns we correlate them with extensive MD simulations, for which retraction of a tip from a gold surface after contact resulted in the formation of a solid gold junction.[4] The elongation of the junction in response to the applied external pulling force, at room temperature, consists of a sequence of stepwise elongation stages. In each elongation stage, atoms in layers (mainly at the vicinity of the narrowest part of the junction) respond first via accumulation of stress accompanied by the occurrence of strained configurations of the wire (which remains ordered in atomic layers, but with increasing nonuniformity of the interlayer spacing). This stage is followed by a shorter atomic disordering and rearrangement period (which may be correlated with the observed dips in the conductance), culminating in the formation of an added layer, with a relief of the accumulated stress and restoration of a higher degree of order in the wire. Consequently, each such elongation necking stage results in a more extended crystalline junction (in increments of the order of the interlayer spacing in the junction, i.e., ~2 Å) of a smaller cross-sectional area [see Figs. 3(a) and 3(b)]. We remark that in these simulations, the overall translation of the tip was performed in an ''adiabatic'' manner allowing for dynamical structural relaxation throughout the elongation process. Failure to allow for such relaxations may result in a sequence of disordered or melted structures.

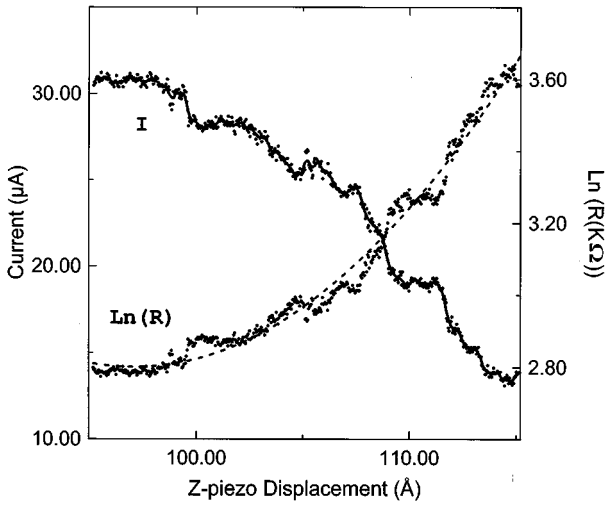

FIG. 1. Plot showing current (left scale) and natural logarithm of the resistance (right scale) vs distance in a 95-Å-long wire as it is pulled 20 Å more until breaking. A voltage of 500 mV was applied in the measurement. The dashed line corresponds to $\ln R=a+b(z-z_0)^2$ with $a=2.8$, $b=2.6\times10^{-3}$, and $z_0=97$ Å.

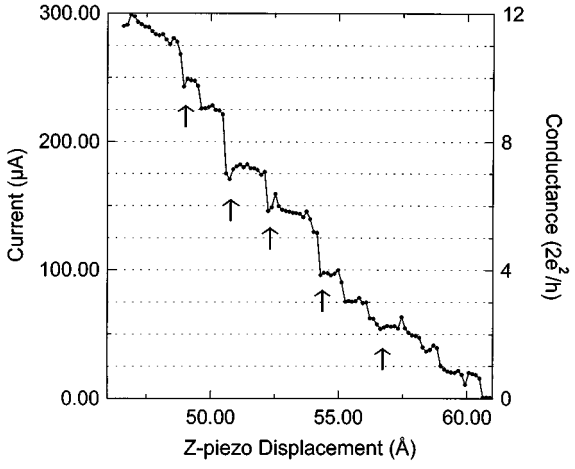

FIG. 2. Current and conductance of a nanowire during an elongation process at room temperature exhibiting conductance quantization steps. Horizontal dashed lines denote intervals of $2e^2/h$. Arrows point to dips interpreted as disordering stages in the elongation process.

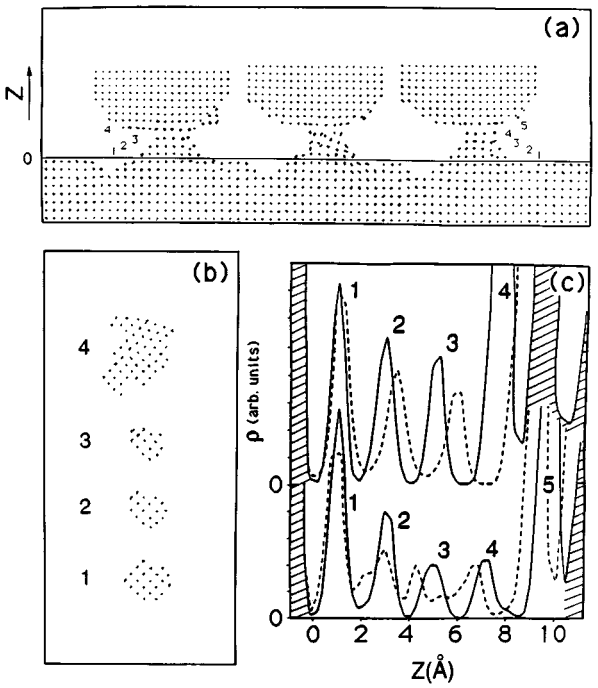

FIG. 3. (a) Side views of atomic configurations obtained from short-time trajectories during a MD simulation of a Ni tip slightly indented into, and then retracted from, an Au(001) surface at 300 K. On the left, a four-layer ordered gold junction formed between the tip and the substrate (the fourth layer of the junction coats the bottom of the tip); the middle configuration demonstrates disorder in the junction during elongation, culminating in the five-layer ordered junction shown on the right. (b) Top views of the in-layer atomic arrangements corresponding to a four-layer ordered wire. (c) Profiles of atomic densities plotted vs distance (Z) along the axis of the wire. The solid lines at the top and bottom correspond to the four-layer and five-layer ordered junctions, respectively [i.e., left and right configurations in (a)]. The dashed line at the top part corresponds to a four-layer strained configuration and the one at the bottom to the disordered structure [middle configuration in (a)] which developed during the elongation process. Hatched regions represent the Au substrate ($Z<0$) and Ni tip layers.

To illustrate the elongation process we show in Fig. 3(a) side views of atomic configurations, starting from a layer-ordered junction containing four atomic layers [see the corresponding intralayer arrangements shown in Fig. 3(b)] and ending with a longer layer-ordered junction containing five layers, along with a structure during the intervening disordered stage. Corresponding plots of the atomic density profiles along the normal axis (Z) of the junction shown in Fig. 3(c) illustrate the atomic distributions in the initial and final ordered stages of the junction as well as during the straining and disordering stages of the transformation.

During the elongation process the wire evolves through atomic configuration with various degrees of order and disorder. We also note that even at the ordered stages, which exhibit crystalline-like atomic layers along the axis of the junction, the shapes of the layers are rather irregular [see Fig. 3(b)], resulting in a solid wire with a surface roughness of a few angstroms, comparable to the wavelength of the electrons (~4 Å). Although such an aspect of disorder may not affect the mechanical characteristics significantly, it can influence electronic transport processes.

A necessary condition for observation of conductance quantization is that the characteristic spacing between energy levels obey $\Delta E > k_B T$, where T is the absolute temperature and k_B is the Boltzmann constant. In semiconductor nanostructures this restricts the experiments to low temperatures. However, in our metallic (gold) nanoscale wires the energy level spacing is relatively large (e.g., for a neck diameter of 10 Å ΔE is of the order of several thousand degrees kelvin). Consequently, temperature effects on conductance quantization in such systems are expected to be negligibly small. Nevertheless the appearance and persistence of quantized conductance in the short wires [see Fig. 2 where nine steps of height $2e^2/h$ or $2(2e^2/h)$, having a period $d \sim 2$ Å, the interlayer spacing in the material, are seen] are quite remarkable, particularly in view of the known sensitivity of conductance quantization to the presence of disorder (which in our wires includes surface roughness, and disorder occurring between the intermittent layer-ordered stages, see Fig. 3).This implies that overall, wires in this length regime maintain a sufficient degree of crystalline order (see Fig. 3) to sustain quantization of the conductance. Furthermore, significant insights into the microscopic mechanism of the elongation process and nature of disorder in the wire are provided by the observation of dips accompanying the quantized conductance steps (see Fig. 2), which can be associated with electron scattering caused by the enhanced structural disorder developing toward the completion of each of the discrete elongation stages of the wire. The rapid rise in the conductance following each dip indicates restoration of a higher degree of order in the pulled wire subsequent to the disordering-rearrangement elongation stage. In addition, the variability in the conductance quantization step height (one or two $2e^2/h$ units) and the above-mentioned occasional occurrence of two successive steps in an elongation interval of combined length $\sim d$ (see Fig. 2) may originate from stick–slip characteristics of the straining–yielding elongation process and from factors influencing the quantization of transverse electronic states (channels) in the wire. The latter include accidental degeneracies of transverse electronic modes, irregular layer shapes, and most likely the occurrence of intermediate atomic configurations during the elongation process which satisfy the condition for closing of a conductance channel. These observations support a correlation between the measured patterns and the aforementioned periodic layerwise order–disorder elongation mechanism of the wire.

We remark that details of the elongation mechanism may depend on the dimensions (length and width) of the wire as well as on the structure, composition, and crystalline orientation of the surface, wire and tip, which can influence the modes of plastic yielding and reordering processes. For example, in the aforementioned simulations involving a Ni tip, a Au(001) surface, and a small radius connective junction-(wire), the elongation process did not appear to involve slip along well-defined glide planes. However, in room-temperature elongation simulations[4(c)] of a longer (75 Å long) and thicker Au(111) wire tapered to a mid-neck radius of ~17 Å, we observed, at various stages, slip events which may occur along a system of glide planes [e.g., (111) and (110)], and more localized (not involving slip) disordering–ordering layer addition processes which, as mentioned

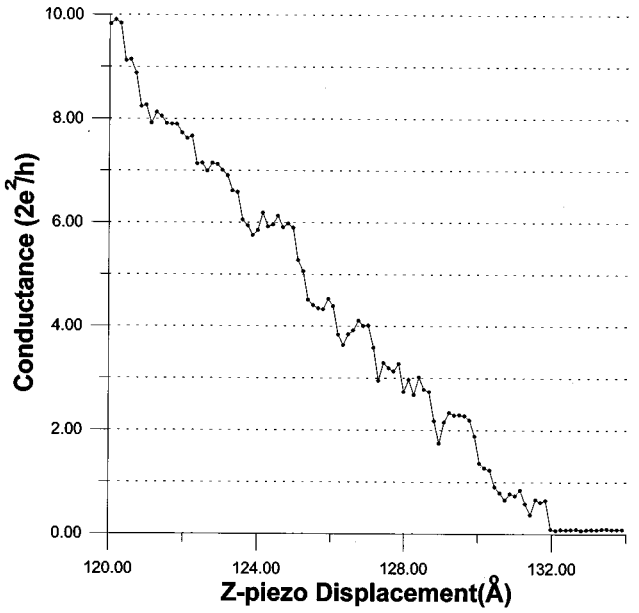

FIG. 4. Conductance vs length for a long and relatively thick wire. The plot shows the typical staircase behavior although the jumps are not so well defined.

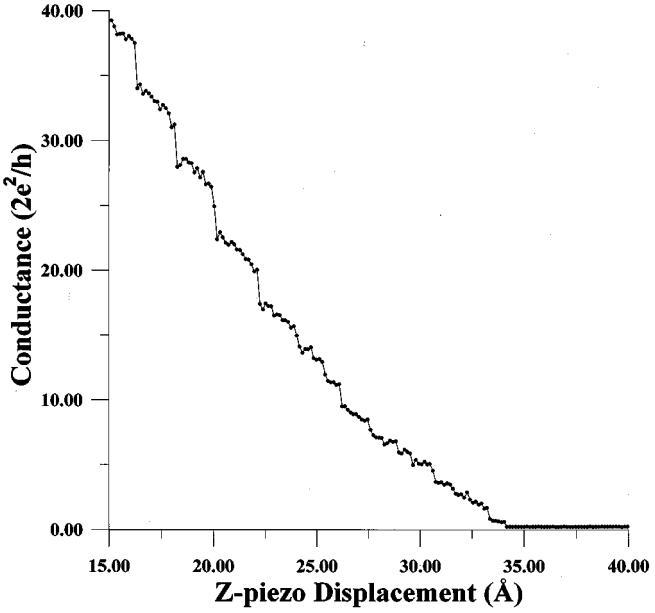

FIG. 5. Plot of the conductance of a short and initially thick wire vs length, showing change in the slope of the plateau between discrete quantization jumps, as the wire is elongated.

above, become dominant for wires of smaller cross sections. Indeed, although we focus here on measurements of conductance quantization in thin wires (i.e., characterized by about 10 or less conductance channels), we note that our experiments on thicker ones show a different pattern (e.g., less well-defined plateaus and steps, and variable conductance step heights), transforming to the behavior shown in Fig. 2 as the wire narrows.

In some wires we have observed effects associated with a relatively higher degree of disorder. This can be seen in Fig. 4 which corresponds to a total elongation of 132 Å. In this thicker wire, localization characteristics were not observed. Rather, the behavior of the resistance can still be interpreted in terms of jumps and dips, but now the values of the conductance steps are not integer multiples of the quantum of conductance, $2e^2/h$, instead they are smaller. This reduction in the conductance per channel can be explained by a decrease in the transmittivity of the channels due to scattering.[3(b),11,12]

The observation of flat plateaus between successive conductance jumps is characteristic to conductance quantization under ballistic adiabatic conditions.[13] When such conditions fail, the plateaus develop negative slopes. As an illustration we show in Fig. 5 results for a thick short wire [note the initial high conductance of $\sim 40(2e^2/h)$]. When the cross section of the wire is large (high conductance), large jumps in the conductance are accompanied by inclined plateaus. As the wire becomes longer and narrower (breaking after an elongation of $\sim$34 Å), the magnitudes of the conductance jumps decrease and the intervening plateaus tend to become flat, indicative of improved quantization conditions in the wire. A similar observation was reported in experiments using mechanically controlled break junctions.[7]

It should be noted that the details of the dependence of the electrical conductance on the wire length vary from experiment to experiment. This indicates that the degree and spatial distribution of disorder in each case, and the particular geometry of the contact, strongly influence the electrical properties of such wires.

In summary, we have shown that metallic nanowires as long as 200 Å can be produced and their electrical properties studied. Different physical phenomena may be observed, depending on the length and the preparation of the wire. In short thin wires, conductance quantization (in units of $2e^2/h$) accompanied by conductance dips[14] occurs (with an approximate periodicity of the interlayer spacing in the wire), and is correlated with a sequential order–disorder mechanism of elongation of the wire, as revealed by MD simulations. In wires longer than the localization length, localization behavior may be observed and $\ln R \sim l^2$. Additionally, we have shown that disorder in such wires and variations in their geometry (which depend on the preparation conditions) may influence the conductance characteristics by reducing the transmittivity of the conductance channels or modifying the quantization conditions.

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