Conductance quantization histograms of gold nanowires at 4 K

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Driving a scanning tunneling microscope (STM) tip into a metallic surface and pulling it out afterwards results in the formation of a nanometer-sized wire (nanowire) between the electrodes. The electrical conduction measured during this process shows signs of quantized conductance in units of $G_0=2e^2/h$. Due to the inherent irreproducibility of the measured conductance curves, the standard technique has been to construct histograms with a few hundred selected curves. These histograms, for gold nanowires at room temperature, have shown three to four peaks at integer values of $2e^2/h$, while in a low-temperature mechanically controlled break junction study, with statistics using only 65 curves, only the first peak has been reported. A proposed explanation for this apparent experimental discrepancy has been a higher nanowire temperature arising from the higher retraction speed used in scanning tunneling microscopy measurements. However, our simple estimation using macroscopic heat transport theory produces a very low temperature rise, less than 1 μK. In this work, an improved statistical study is presented, where histograms built with thousands of consecutive gold contact breakage experiments at 4 K show up to seven conductance peaks. Thus, no significant differences with our previous room-temperature (RT) studies are observed, pointing to a conductance quantization behavior that is the same at low, intermediate, and high (RT) temperatures. [S0163-1829(97)04619-5]

The electrical conductance through a narrow constriction, having a diameter of the order of the electron wavelength, is quantized in units of $G_0=2e^2/h$.[1,2] Such conductance quantization has been observed in semiconductor devices containing a two-dimensional electron gas at low temperature.[3] Similar effects are possible[4] at room temperature in metallic wires with a diameter of the order of 1 nm (nanowires) and have been observed using scanning tunneling microscopy (STM),[5,6] mechanically controlled break junctions (MCBJ),[7–9] and, as shown recently,[10,11] just using plain household wires. These techniques use the same basic principle: by pressing two metals together, a metallic contact is formed that can be stretched to a nanowire by the subsequent separation of the electrodes. The conductance in such a system is found to decrease in abrupt steps, of about $2e^2/h$ height, just before the contact breaks. Due to the inherent irreproducibility of the measured conductance curves, histograms have been built using a few hundred experimental curves.[12] The scientific relevance of these histograms has been the subject of recent controversy,[12,13] concerning whether the stepped behavior of the conductance is due to conductance quantization, or to discrete contact size changes during the nanowire elongation process due to the discrete atomic size.

In a MCBJ study by Krans[9] on Au nanowires at 4.2 K, no sign of conductance quantization is observed apart from a well-defined conductance peak at $1G_0$, while at 77 K three conductance peaks are observed. In a recent MCBJ study, Muller *et al.*,[8] showed that, at room temperature, conductance quantization is observed more prominently than in the earlier low-temperature measurements. This agrees with all STM experiments on gold at room temperature.[5,6] The explanation given[8,9] for this temperature dependency in the MCBJ experiments was that for higher temperature the contact can explore a wider range of structures to find the most favorable atomic configuration, due to the higher kinetic energy of the atoms. This allows the nanowire to find a more relaxed form, and quantized conductance is observed. To some extent these considerations are also supported by numerical simulations by Bratkovsky, Sutton, and Todorov,[14] where the nanowire tends to become more uniform in cross section, and internal defects are seen to vanish more easily at high temperatures. However, the relevance to the experiments is not quite clear, since the pulling speed is many orders of magnitude larger in the simulations than in the experiments (3.5 m s^{-1} for simulations, 1 μm s^{-1} for STM experiments, and 0.01 nm s^{-1} for the MCBJ experiments). In contrast to MCBJ experiments, a recent STM study[15] by Sirvent *et al.* shows three clear conductance peaks, well resolved at three different temperatures: 4, 77, and 300 K and with no significant difference between the histograms obtained at these three temperatures. These peaks are not located exactly at integer values of G_0 and there is no discussion of the possible origin of this experimental observation.

In this paper we show that by using a STM at 4 K, up to seven conductance peaks are resolved. This improves previous studies in the sense that (i) no selection criteria are used to select the conductance curves with which the histogram is built, (ii) up to five peaks fall on integer values of G_0 upon the subtraction of a constant resistance, (iii) disorder is presented as a good candidate to explain this histogram peak shift, and, (iv) the histograms obtained using a few thousand conductance curves are shown to be totally reproducible. Furthermore, using a simple estimation based on macro-

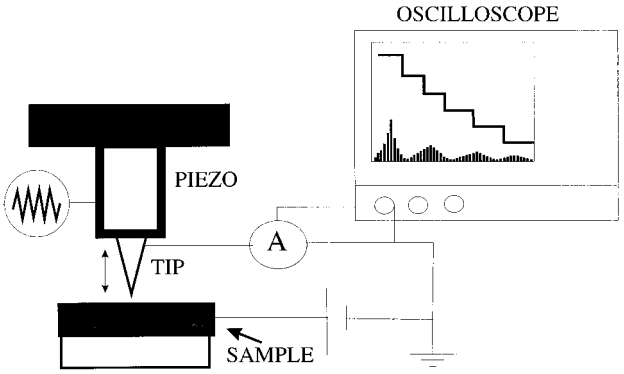

FIG. 1. Schematic drawing of our experimental setup: A gold tip is crushed repeatedly onto a gold surface at 4 K driving the tip piezoelectric with a triangular signal. The conductance is measured at the last stages of the contact breakage process. The histogram is built with all the measured curves and is displayed in real time.

scopic heat transport theory, the suspicion that the higher separation speed in STM as compared to MCBJ will raise the nanowire temperature is discarded. This effect could have explained the proposed experimental discrepancies between MCBJ and STM histograms at low temperature.

The STM tips are made of 0.25-mm 99.99% pure gold and the samples are evaporated gold thin films. The measurements are carried out in vacuum using a low-temperature STM at 4 K. A triangular waveform is fed to the piezoactuator, driving repeatedly the tip in and out of contact with the sample surface, with a typical frequency of 5 Hz and an amplitude of 1.4 μm, i.e., a 14-μm s^{-1} speed. The speed is modified changing the amplitude and/or frequency of this signal. During the electrodes' approach and separation the conductance is measured using a current to voltage converter with 10^5 gain (100 mV/μA), 1 μs rise time, and 3 μs settling time. A 500-MHz bandwidth and 5-G samples/s, 8-bit, digital oscilloscope (LeCroy 9345AM) is used to build the histograms in real time. The tip-sample bias in all experiments reported here is 90.3 mV, corresponding to a quantum current step of 7 μA ($2e^2/h$ is about 77.5 μS). Notice that all the measured curves are used to build the histogram. A schematic drawing of our experimental setup is shown in Fig. 1. In this work we present histograms of current decays, i.e., only contact breakage experiments are used to construct the histogram. This histogram is corrected for the nonideal differential linearity (NIDL) of the oscilloscope.[16] This problem can be described as follows: When the histogram bin size is selected to accommodate an integer number of digital levels, the histogram of a continuously decreasing straight line should be flat but it is not. This histogram, normalized to one, is shown in Fig. 2(c) below. Previous to this work, we smoothed the obtained histograms averaging two or three bins to the side, the amount that produced a flat histogram when measuring straight lines. Both approaches obtain a similar result. However, the NIDL correction produces sharper peaks because it is not a smoothing procedure, it eliminates a systematic error. This correction is performed by dividing the raw histogram over the normalized NIDL, measured under the same conditions and in the same channel of the oscilloscope as the conductance data.

Figure 2(a) shows the raw conductance histogram data, for a series of 5 000 consecutive indentation experiments using Au electrodes at 4.2 K and with an electrode speed of about 20 μm s. Figure 2(b) shows the data corrected for the NIDL. The measured NIDL signal, normalized, is shown in Fig. 2(c). We observe in Fig. 2(b) at least five peaks, slightly separated from integer values of G_0. The histogram shown in Fig. 2(b) is displayed in a vertical logarithmic scale in Fig. 3(a) in order to magnify the detail. Notice that up to seven conductance peaks are resolved. In Fig. 3(b) the conductance values are displayed after subtracting a series resistance of 390 Ω. This has been a common procedure in all quantized conductance reports, and has been justified by the resistance of the leads, i.e., what is not the nanowire itself. As shown recently in a theoretical work, this resistance could arise also from disorder within the nanowire.[17] Recent experimental results, including the ones presented here, also support this

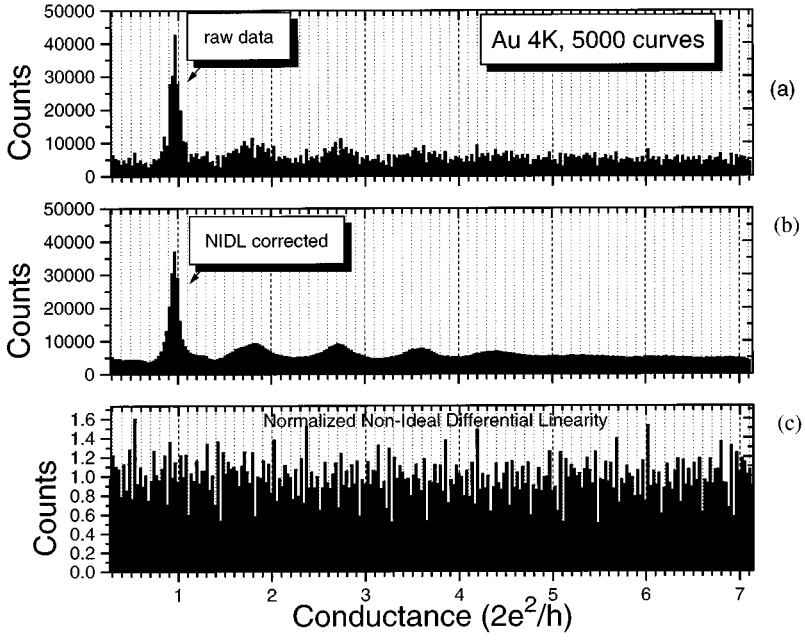

FIG. 2. Conductance histogram for gold nanowires at 4 K built with 5 000 consecutive curves. The applied bias between tip and sample is 90.4 mV, the tip retraction speed is 20 μm s^{-1}. (a) displays raw data; (b) shows the data corrected for the non-ideal differential linearity of the oscilloscope (NIDL); (c) shows the normalized NIDL of the oscilloscope channel, measured under the same conditions as the conductance experiments.

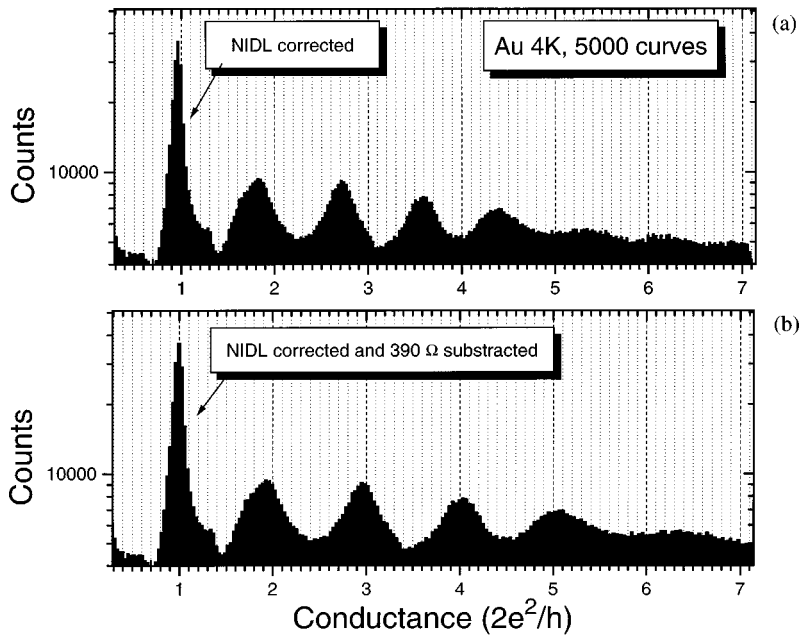

FIG. 3. The data shown in Fig. 2(b) are displayed in (a) in a vertical logarithmic scale in order to magnify the peak structure. Notice that up to seven peaks are resolved. (b) displays the result of substracting 390 Ω to the conductance. Notice that the first five peaks fall on integer values of $G_0=2e^2/h=1/12\,907\ \Omega$.

view. Figure 4 shows a histogram built with 900 000 consecutive curves, corrected for the NIDL, with a very similar appearance as that in Fig. 3(b). The electrode separation speed was also 20 μm s^{-1} and a series resistance of 390 Ω has been subtracted. Note that this histogram proves the good statistical behavior of the gold nanowire conductance and the reproducibility of the experiment. The inset shows a typical conductance curve.

In the histograms shown in Figs. 2, 3, and 4 as the conductance value increases, the peak height decreases and the width increases, in agreement with room-temperature (RT) STM experiments.[5,6] This also agrees qualitatively with recent theoretical calculations;[6] however, in our experiments, we can resolve peak structures well beyond the fourth conductance peak as shown in Figs. 2, 3, and 4. In our RT studies[10,11] we find this behavior as well. The above experimental facts are in contrast with MCBJ experiments[8] at RT, which show peaks with almost the same height-width ratio. However, the main difference between the present results and the MCBJ results at 4 K is that the MCBJ histogram only shows a well-defined peak at $G_n=1G_0$. Besides poor statistics, a proposed explanation for this discrepancy has been that the temperature in the wire is not the same as in the surrounding bath, due to the higher separation speed used in our STM experiments. In the MCBJ experiments the separation speed is orders of magnitude lower than in our case (0.1 nm s^{-1} in MCBJ, and, 10 000 nm s^{-1} in STM experiments). If the energy released during the elongation process is transformed into heat, and this heat is not transported away rapidly enough, the temperature in the wire will be higher than

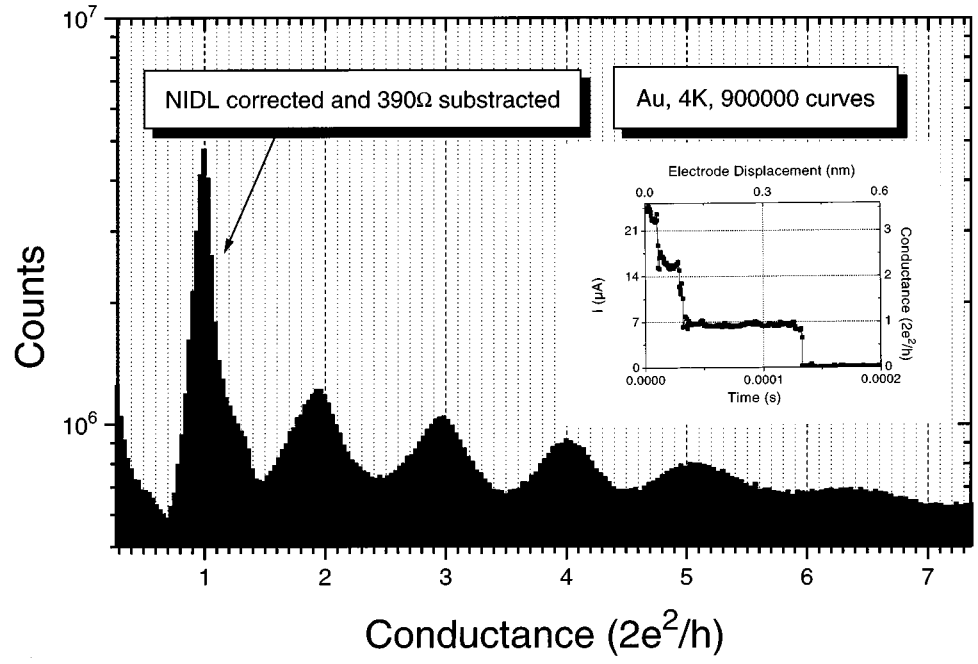

FIG. 4. Conductance histogram built with 900 000 consecutive contact breakage curves shown in a vertical logarithmic scale, with the NIDL correction and 390 Ω substracted. The experimental conditions are identical to the ones mentioned in Fig. 2. The inset shows a typical conductance versus time staircase.

in the surrounding bath. A higher temperature will produce a more favorable wire from the quantized conductance point of view, as discussed by Krans and co-workers[8,9] and in the theoretical work by Bratkovsky, Sutton, and Todorov.[14] They show that at high temperatures the nanowire tends to become more uniform in the cross section and internal structure defects disappear more rapidly.

A simple estimation of the temperature increase could be obtained if one considers the amount of heat that is released during the pulling of the wire. The heat within the nanowire arises from the energy released during the bond-breaking process. This heat is transported away from the nanowire through the reservoir. We can estimate the temperature increase of the nanowire, with respect to the reservoir if the power generated during the wire elongation is known. The inset in Fig. 3 shows a typical conductance versus time graph, where the tip retraction speed is 3.1 μm s^{-1}. As observed, the lifetime of the last conductance plateau is about 100 μs. During the pulling process, the contact cross section decreases, surface area increases, and energy is released. If the cross-sectional area of the wire S is reduced by a factor of 2, the surface area will increase by a factor of $2^{0.5}=1.41$ under constant volume conditions. If the wire is a cylinder with 1 nm diameter and 2 nm length, the initial surface area will change from 6.3×10^{-18} to 8.8×10^{-18} m^2. Considering a typical area of an atom of $(2\times10^{-10})^2$ m^2, and a typical difference between binding energy in the surface and in the bulk of 1 eV per atom, a total energy of 62.5 eV will be released during such an elongation. If these 62.5 eV are released during this last plateau duration, 100 μs, the power generated will be about 0.1 pW. One may get a simple estimation of the temperature increase in the nanowire using Fourier's law for thermal conduction, considering that this power is transported away from the nanowire through the reservoir,

$$j=-\kappa\,\delta T/\delta r \quad (\mathrm{J\ s^{-1}\ m^{-2}}), \tag{1}$$

where j is the thermal current density, κ the thermal conductivity, and $\delta T/\delta r$ is the temperature gradient. Integrating the equation, the temperature increase can be expressed as

$$\Delta T = W/2\pi\kappa a \quad (\mathrm{K}), \tag{2}$$

where $W=Q/\Delta t=0.1$ pW is the power dissipated, and a is the nanowire size. Assuming that κ for Au at 4 K is about 1000 W mK typical for a high-purity material, and a is about 1 nm, the temperature increase is less than 1 μK. Even if κ or the dissipation time are decreased orders of magnitude this estimation will still produce a very low temperature increase in the nanowire.

Summarizing, a conductance histogram with five clear peaks at integer values of the quantized conductance value is observed in gold nanowires at 4 K. The height of the conductance peaks decreases and the width of the peaks increases with increasing conductance, in agreement with room-temperature STM experiments.[6,11] It has been argued that the higher speed used in our STM experiments as compared to MCBJ experiments might result in a higher nano wire temperature. However, a simple estimation, based on macroscopic heat transport, produces a temperature rise in the nanowire of the order of μ K, ruling out this explanation. Therefore, our improved statistical study shows that conductance quantization in gold nanowires is basically the same at high, low, and intermediate temperatures.

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