Conductance of atomic-scale gold contacts under high-bias voltages

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The conductance of atomic-scale gold contacts has been studied experimentally as a function of bias voltage. At low voltages, the conductance histogram shows a pronounced peak at $G_0=2e^2/h$ and broad peaks near $2G_0$ and $3G_0$. With increasing the bias voltage, these peaks decrease in height and disappear at some critical voltage. The $1G_0$ peak is found to disappear at 1.9 V at room temperature, which corresponds to a critical current of 137 μA. At liquid-nitrogen temperature, the critical voltage and current slightly increases to 2.2 V and 165 μA, respectively. The observed disappearance of the conductance peaks can be interpreted as a contact instability due to the electromigration of contact atoms induced by a high current density.
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In the past few years, there have been a number of experimental studies of conductance quantization in metallic quantum point contacts using scanning tunneling microscopy,[1–6] mechanically controllable break junctions,[7,8] and a momentary touching of macroscopic electrodes.[9,10] In these experiments, an atomic-size constriction is formed by a pulloff of two electrodes in contact. A connective neck between electrodes is stretched out until it consists of only a few atoms. Conductance plateaus and steps at and near the integer multiples of $G_0=2e^2/h$ are observed in this last stage of contact break. Recent experimental and theoretical studies[5,6,11–14] have revealed that these steps are primarily due to discrete atomic rearrangements in the contact. Since the contact atoms do not rearrange themselves in a unique way, the plateaus and steps are not reproducible. A conductance histogram[3,4,7–9] is thus generally used to represent the conductance properties of the contacts. Peaks appear in the histogram when the contact has stable atomic geometries and exhibits conductance plateaus. Each conductance peak is exactly positioned at nG_0 only if the corresponding contact geometry makes no electron backscattering.

In previous experiments, the voltage applied to the contact is typically 100 mV or lower and the contact current at the $1G_0$ plateau is less than 10 μA. Ideally, it would be possible to increase the contact current until it reaches a ''space-charge limit'' put by electron correlations at the contact. When the contact consists of one atom, this current limit can be estimated as $I_m \sim ev_F/a$, where v_F is the Fermi velocity and a an atomic distance. For a gold contact, $I_m \sim 770$ μA. Since the single-atom conductance is 1 G_0, this current would generate a bias voltage of 10 V if the conductance continues to be quantized to $1G_0$. In practice, the quantum conductance under such a high voltage is quite unlikely and the breakdown of the conductance quantization would be expected at lower voltages.

In this paper we report our conductance measurements on atomic-size gold contacts under high voltages. Our experiment is similar to the wire-separation experiment of Costa-Krämer *et al.*[9] Instead of wires, however, we have used the gold-coated relay contacts which have been found to exhibit clear quantized conductance steps during their contact break.[15] To measure the relay conductance, a current-sensing resistor (998 Ω) is connected in series with a relay and a dc voltage is applied to the circuit. A voltage drop across the resistor is monitored with a fast digital oscilloscope and converted to the relay conductance. Note that in our experimental setup, the bias voltage of the relay contact should be distinguished from the dc applied voltage to the circuit. The bias voltage varies with the relay conductance and becomes 7% lower than the applied voltage when the relay conductance is $1G_0$.

Figure 1 shows the transient conductance trace measured on a gold-contact relay (OMRON G5V-1) under an applied voltage of 500 mV at room temperature. The trace clearly shows marked plateaus near integer multiples of G_0. As is usually observed in metal contacts, the conductance trace has little reproducibility and differs for each contact break. Nevertheless we find in average that the last $1G_0$ plateau appears most frequently and has a longer width. This plateau also gives a better coincidence with $1G_0$, while higher plateaus tend to deviate from their quantized values as seen in Fig. 1. The tail at each step is due to the finite time resolution of our measuring system. The transition between the plateaus is abrupt as indicated by the sharp upper edge of each step.

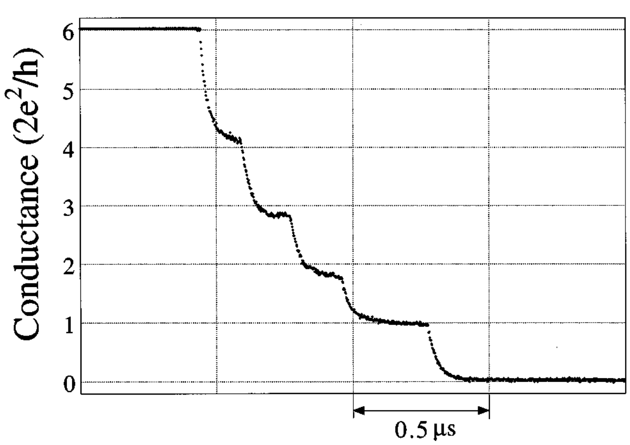

FIG. 1. Transient conductance trace in the contact break of a gold-contact relay observed at room temperature under an applied voltage of 500 mV. A tail at each step is due to the finite time constant of our measuring system.

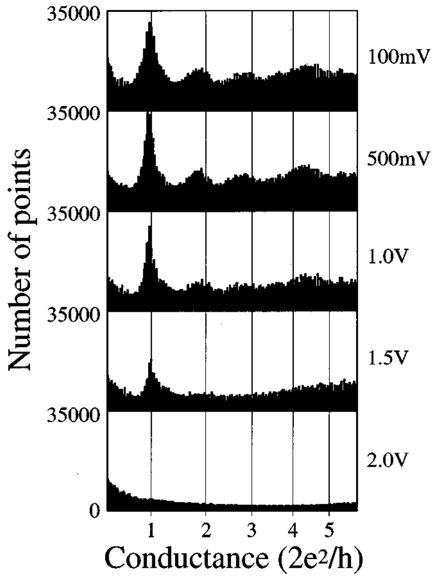

FIG. 2. Conductance histograms for a gold-contact relay obtained at various applied voltages. Each histogram is constructed from 10 000 conductance data. Note that for the $1G_0$ peak, the bias voltage is 7% lower than the applied voltage indicated in the figure.

We have recorded conductance traces at various applied voltages and constructed the conductance histograms shown in Fig. 2. Each histogram is constructed from 10 000 traces. Two histograms obtained at 100 and 500 mV are almost identical. They show a pronounced peak at $1G_0$ and smaller peaks near $2G_0$, $3G_0$, and around 4 G_0. The first three peaks are positioned at 0.95 G_0, $1.8G_0$, and 2.7 G_0, respectively. These histograms are in good agreement with the reported histograms for gold quantum contacts,[3,8,9] in particular with the one obtained by Costa-Krämer *et al.*[16] The position of the first peak also coincides with the reported value (0.98 G_0) for the last plateau in gold contact.[4,5] When the applied voltage is increased to 1.0 V, the $2G_0$ and higher peaks start to decrease. They all disappear at 1.5 V. The $1G_0$ remains almost unchanged at 1.0 V but decreases at 1.5 V. Finally at 2.0 V, the histogram shows no peak structures. We find that the change of histogram shown in Fig. 2 is quite reproducible. When the applied voltage is reduced from 2.0 V to 100 mV, the histogram returns to the top one in Fig. 2. This confirms that the contact suffers no permanent damages through operations at 2.0 V. The same voltage dependence as shown in Fig. 2 is also found in the histograms obtained on other gold-contact relays (another G5V-1 and MATSUSHITA DS1E). Therefore the disappearance of the conductance peaks at high voltages is not a specific property of the relay investigated but a general characteristic of gold contacts.

The histograms in Fig. 2 reveal an essential difference between the conductance of our gold contacts and that of ballistic contacts in a two-dimensional electron gas (2DEG). In the 2DEG quantum point contacts, the conductance quantization breaks down when the voltage becomes comparable to the subband separation.[17] Above this critical voltage, the conductance becomes nonlinear and deviates from nG_0. Also the conductance steps are smeared at high voltages due to various broadening mechanisms.[18] None of these effects

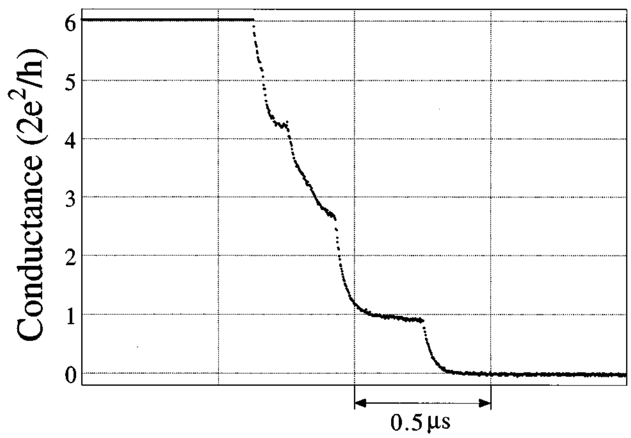

FIG. 3. Conductance trace in a contact break under an applied voltage of 1.5 V. The $1G_0$ step shows no smearing.

are observed in our experiment. In Fig. 2, conductance peaks show no appreciable shift of their position with increasing the applied voltage; only their peak height is reduced. Also no new peaks appear in the histogram. Figure 3 displays a conductance trace recorded under an applied voltage of 1.5 V. Comparison of it with Fig. 1 clearly shows that the $1G_0$ plateau at 1.5 V has a well-defined step and suffers no broadening effects. We can conclude from these observations that the peak disappearance in Fig. 2 is not due to the high-voltage broadening mechanisms in the 2DEG quantum point contacts.

In order to determine the critical voltage for the disappearance of the $1G_0$ peak, we have measured the conductance with a 0.1-V step at room temperature and also at a liquid-nitrogen temperature by directly dipping the relays into liquid nitrogen. Figures 4(a) and 4(b) show the conductance histograms obtained at 300 K and at 77 K, respectively. It can be seen in Fig. 4(a) that the $1G_0$ peak at room

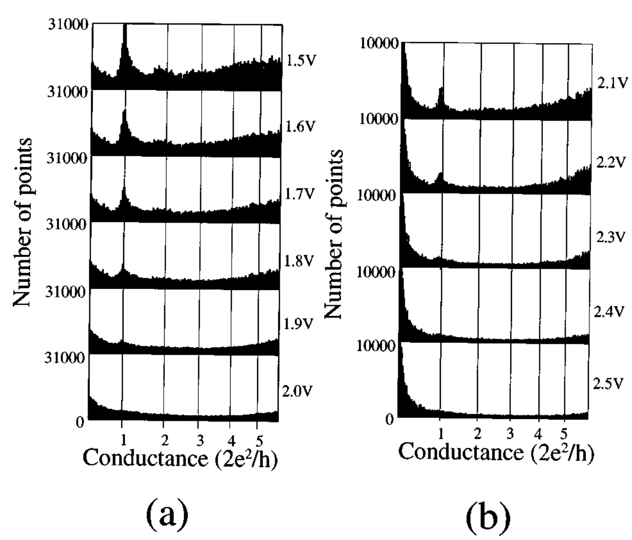

FIG. 4. Disappearance of the $1G_0$ peak with increasing the applied voltage (a) at room temperature and (b) at liquid-nitrogen temperature. Histograms in (a) and (b) are constructed from 10 000 and 2000 traces, respectively. In each histogram, the bias voltage for the $1G_0$ peak is 7% lower than the applied voltage as in Fig. 2.

temperature gradually decreases in height with increasing the applied voltage and disappears at 2.0 V. In other gold-contact relays, we find this critical voltage to be 1.9–2.2 V. At liquid-nitrogen temperature, the $1G_0$ peak well exists at 2.2 V but disappears at 2.4 V [Fig. 4(b)]. Upon heating back to room temperature, the $1G_0$ peak again disappears at 2.0 V. This result assures that the increase in the critical voltage at 77 K is due to sample cooling. Note that the bias voltage of the $1G_0$ contact is 7% lower than the applied voltage. The critical *bias* voltage thus becomes 1.9 V at 300 K and 2.2 V at 77 K. At these critical voltages, the current flowing through the $1G_0$ contact is calculated to be 137 and 165 μA, respectively. it is generally believed that the $1G_0$ contact consists of just one atom.[5,7,12] Our experimental results then indicate that one gold atom can conduct a current as high as 165 μA. This current is of the same order of magnitude as the current limit I_m. In this sense, it can be said that the $1G_0$ contact is close to the ideal single-atom contact.

Since the current density in the contact becomes extremely high ($\sim 10^{11}$ A/cm^2) at the critical voltage, we interpret the observed disappearance of conductance peaks as being due to a current-induced contact instability caused by the self-electromigration of contact atoms. The driving force for electromigration has two components: the direct field force and the electron-wind force.[19] Since the direct force vanishes in one-dimensional conductors,[20] we here consider only the electron-wind force due to momentum transfer from electrons to atoms. The wind force acting on the contact atoms can be written as[21]

$$F_w = 2(Ja^2/e) m v_F \eta,$$

where J is the current density, a the atomic distance, m the electron mass, and v_F the Fermi velocity. A constant η depends on the scattering geometry and takes values between 0 (forward scattering) and 1 (backscattering). If we assume a single-atom contact and substitute for Ja^2 the experimental critical current, then we obtain for the maximum wind force $F_{\rm wc} \sim 2.2$ nN at room temperature and $F_{\rm wc} \sim 2.6$ nN at nitrogen temperature. These values well exceed the breaking force 1.5 nN of the single-atom contact of gold.[5,14] The actual strength of the wind force, however, would take smaller values depending on the contact geometry and the coefficient η in Eq. (1). If the conductance is perfectly quantized, for example, the wind force vanishes since $\eta = 0$. Perhaps such a mechanism is responsible for the long-lived $1G_0$ plateau seen in Fig. 3. On the other hand, the $2G_0$ and higher plateaus which often deviate from nG_0 disappear at lower voltages as expected. The wind force is thus not dominant once a stable atomic geometry with low η is formed in the contact. However, during the atomic rearrangements prior to the formation of stable geometries, the wind force is likely to be enhanced since this process involves substantial atomic disordering[12,13] which gives rise to increased electron scattering. The strong electromigration force during atomic rearrangements would promote the directive motion of contact atoms and suppress the relaxation of atoms into stable geometries. This explains the decrease of the conductance peak height with increasing the bias voltage. At the critical voltage, the contact would have no chance of forming stable atomic geometries and undergo continuous deformation. In terms of the motion of individual contact atoms, this instability can be viewed as the collapse of the migration barrier due to the electromigration force. An expression for the effective activation barrier modified by electromigration is $\epsilon - \zeta V_v$, where V_b is the bias voltage, ϵ the energy barrier at zero bias, and ζ a parameter describing the contribution of electromigration.[21,22] The instability takes place when the effective barrier becomes comparable to the thermal energy, $\epsilon - \zeta V_{\rm bc} = k_B T$, where $V_{\rm bc}$ is the critical bias voltage. This relation accounts for the observed increase in $V_{\rm bc}$ at low temperature. If we take $\zeta \sim 0.1$ eV/V as a representative value for noble metals,[21] then we can estimate $V_{\rm bc}$ at 77 K from its experimental value at 300 K. The result is $V_{\rm bc} \sim 21$ V in good agreement with the observed $V_{\rm bc}$ of 2.2 V at 77 K. Such a numerical coincidence may be fortuitous considering the crudeness of our approach and the large uncertainty in ζ.[21,22] We note however that the critical electromigration pressure $\zeta V_{\rm bc}/a^3 \sim 1.3$ GPa is comparable to the mechanical strength of gold nanowires.[5,10] Therefore, the parameter $\zeta \sim 0.1$ eV/V may be a reasonable measure of the electromigration force during the plastic flow in the contact.

Finally, we briefly discuss the effects of Joule heating. In ballistic contacts, electrons dissipate their acquired energy in the ''exit'' region connected to the constriction. This local heating however critically depends on the size and the thermal conductivity of the exit region, both of which are totally unknown. If we use the electron mean free path of bulk gold for the size of the exit region and assume the Wiedemann-Franz law to calculate the thermal conductivity, then we obtain $\Delta T \sim 76$ K for the temperature rise of the $1G_0$ contact at the critical voltage at 300 K and $\Delta \dagger T \sim 16$ K at 77 K. The contact temperature would be higher if the electron mean free path is limited by some atomic disordering in the contact. However, molecular-dynamics simulations[13] suggest that the increased thermal motion of contact atoms tends to promote the relaxation of atoms into stable ordered structures. The pure thermal instability due to Joule heating alone is thus unlikely unless the contact temperature becomes close to its melting point.

In summary we have studied the conductance of atomic-scale gold contacts under high-bias voltages. Our results show that the increase in the bias voltage produces the contact instability and decreases the conductance peaks in the histogram. This peak disappearance has no relation to the conventional broadening effects in quantum point contacts. We find that the electromigration of contact atoms due to high contact current gives a reasonable account of the observed contact instability under high-bias voltages.

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