Stability of Atom-sized Metal Contacts under High Biases*

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Conductance and its bias dependence have been measured on Au and Ag breaking nanocontacts for biases up to 3.0 V at room temperature in ultrahigh vacuum. Under high-bias/high-current conditions, both Au and Ag contacts often show a characteristic conductance fluctuation when the conductance attains a certain critical value $G_{th}$. This critical conductance ranges from $\sim 10G_0$ to $\sim 50G_0$ ($G_0 \equiv 2e^2/h$ is the quantum unit of conductance) and increases with the bias. When $G_{th}$ is plotted against the contact current $I$, we obtained a linear $I-G_{th}$ plot for Au contacts. Since $G_{th}$ is in a semiclassical regime and hence should be proportional to the cross sectional area of the contact, the slope of the $I-G_{th}$ plot represents a critical current density for the onset of the conductance fluctuation. These observations indicate that the conductance fluctuation is due to certain current-induced contact instability. When the current density exceeds the critical value, a contact becomes unstable and tends to be ruptured, leaving small chances of further necking deformation down to a single-atom contact. [DOI: 10.1380/ejssnt.2004.125]

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I. INTRODUCTION

Electron transport through metal nanocontacts has been a subject of intensive experimental and theoretical investigations [1]. Particularly interesting is a single-atom contact (SAC) which bridges two electrodes through a single atom or a single chain of atoms. Previous studies have revealed that the SAC takes a characteristic conductance, the value of which is element specific. In the case of noble metal SACs, their ballistic conductance agrees quite well with the quantum unit of conductance $G_0 \equiv 2e^2/h$. Because of this universal and material-independent conductance, these SACs would be a potential candidate as a wiring material in atomic and molecular devices.

In most experiments, a metal nanocontact is studied using a breaking junction technique. When two electrodes in contact are pulled apart, the contact stretches out and forms a connective neck between electrodes. At the last stage of junction break, the narrowest constriction shrinks to the size of atoms and ultimately becomes a SAC. Unfortunately, we have at present no control over the atomic-scale necking deformation of the contact, and as a result, SACs can be formed only by chance. In this sense, the SAC formation is a ‘statistical’ event. We, therefore, have to repeat breaking junctions many times and acquire a large amount of data. Properties of SACs can be deduced by statistically analyzing these data. Usually, we monitor transient conductance during contact break and can detect the SAC formation by observing a conductance plateau, which occasionally appears just before the contact break. If such a plateau appears at a conductance value specific to SAC, it unambiguously signals the SAC formation. Because of the statistical nature of the SAC formation, not all but finite fractions of transient conductance traces display such SAC plateaus. Nevertheless, by counting the number of SAC plateaus, we can obtain the formation probability $p$ of SAC. Since the conductance of Au and Ag SACs is $1G_0$, we can obtain their formation probabilities $p_{Au}$ and $p_{Ag}$ from the number of $1G_0$ plateaus in their conductance traces.

Among metal SACs, Au SACs appear to have superior stability since they can be observed under various environments. In our previous experiments, we measured the high-bias conductance of Au [2,3,4] and Au alloys [5], and studied how $p_{Au}$ changes with the bias voltage. Our experimental results showed that $p_{Au}$ decreases with increasing the bias and vanishes around 2 V. This critical bias nicely corresponds to 2.3 V at which almost all $1G_0$ contacts of Au become disrupted [6]. These results indicate that Au SACs are highly stable against high biases and capable of holding a $\sim 2$ V voltage drop or sustaining a contact current of $\sim 150$ $\mu$A. Compared to Au SACs, Al SACs are found to be less stable. Our conductance measurements on Al SACs [4] showed that $p_{Al}$ vanishes at 0.8 V, well below the critical bias of $p_{Au}$. It, however, remains unclear why Au SACs survive up to high biases while Al SACs do not.

In this work, we measured the high bias conductance of Ag nanocontacts and investigated the bias dependence of $p_{Ag}$. We took up Ag SACs because Au and Ag SACs have nearly the same electronic properties, and the comparison of their formation probabilities is expected to give some clue for understanding the stability of metal SACs against high biases. We also present in this paper our new finding of a rapid conductance fluctuation in Au and Ag nanocontacts. This fluctuation usually takes place when the conductance of a breaking contact becomes $10 \sim 50G_0$ and...

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with the tip-disk contact. We recorded a voltage drop across a current-sensing resistor \( R_0 \) connected in series with the tip-disk contact. Actually, we did not directly measure the conductance but monitored a transient conductance during contact break as mentioned in Sec. I, we detected the SAC formation by monitoring a transient conductance during contact break with a fast digital oscilloscope (Tektronix TDS3052). Actually, we did not directly measure the conductance but recorded a voltage drop \( V_m \) across a fixed resistor \( R_0 \) (1 k\( \Omega \)), which was connected in series with the contact as shown in Fig. 1. The contact conductance \( G \) was calculated through a formula,

\[
G = \frac{V_m}{R_0(V_a - V_m)}. \tag{1}
\]

where \( V_a \) is an applied bias voltage supplied from a DC source (KEITHLEY 2400). A true bias voltage \( V_b \) across the contact is not \( V_a \) but \( V_b = V_a - V_m \) and is given as,

\[
V_b = \frac{V_a}{1 + GR_0}. \tag{2}
\]

The contact current \( I \) is related to \( V_a \) as,

\[
I = \frac{V_aG}{1 + GR_0}. \tag{3}
\]

Therefore, the contact current depends nonlinearly on \( G \). Since the SAC formation is a statistical event, we have to accumulate a large number of conductance traces. In this experiment, we repeated the contact ON-OFF at 1 Hz and recorded 2000 traces for each bias. For Au contacts, we changed the applied bias from 0.4 V to 3.2 V, while for Ag contacts, we had to limit the bias below 1 V and employed a bias range spanning from 0.1 V to 0.8 V, since \( p_{Ag} \) almost vanishes at 0.6 V as we will see in the next section. For detecting SAC conductance plateaus, we usually monitor a conductance trace in a conductance range from 0 to 4\( G_0 \). In some measurements, however, we extended the monitor range up to 100\( G_0 \) for examining behaviors of conductance traces in their high-conductance regime.

All measurements were carried out at room temperature in UHV with a base pressure of \( \sim 2 \times 10^{-8} \) Pa.

**III. RESULTS**

**A. Formation probability of \( 1G_0 \) plateaus and its bias dependence**

Figure 2 shows a typical conductance trace of Au and Ag contacts obtained during a contact break at 0.4 V and 0.2 V, respectively. As mentioned before, only a part of each conductance trace below 4\( G_0 \) was measured and displayed in the figure. In both traces, the conductance decreases stepwise and shows a plateau at 1\( G_0 \). Because of lower S/N ratio at 0.2 V, the trace in Fig. 2(b) exhibits larger noises than the one shown in Fig. 2(a). Nevertheless, the 1\( G_0 \) plateau can be well identified. Since both Au and Ag SACs have a specific conductance of 1\( G_0 \), these 1\( G_0 \) plateaus certainly indicate the formation of Au and Ag SACs.

The number of these 1\( G_0 \) plateaus gives us the formation probabilities \( p_{Au} \) and \( p_{Ag} \) of Au and Ag SACs, but to begin with, we have to discriminate genuine 1\( G_0 \) plateaus from noises. This discrimination is by no means trivial since short plateaus are hardly distinguishable from noises. In this work, we ‘defined’ the 1\( G_0 \) plateau as those plateaus which exist in the conductance range 0.9 \( \sim 1.1G_0 \) and survive longer than 2 \( \mu s \). With this criterion, we counted the number of 1\( G_0 \) plateaus and obtained the SAC formation probability by dividing it by the number of recorded conductance traces (2000 in this work). Note that the formation probability thus obtained is the one for SACs having a lifetime longer than 2 \( \mu s \). Short-lived plateaus could be detected by zooming up the time scale of the oscilloscope, but long plateaus would then be missed. With a limited recording memory of the oscilloscope, we cannot record both short and long plateaus, and therefore, have to make some compromise.
FIG. 2: Typical conductance traces of (a) Au and (b) Ag breaking contacts recorded at 0.4 V and 0.2 V, respectively. In both traces, the conductance decreases stepwise and shows a 1G₀ plateau.

We plot in Fig. 3 p_Au and p_Ag as a function of the bias voltage. Both p_Au and p_Ag decrease with increasing the bias, and their plots are rather similar in shape. However, their relevant bias range is quite different. The observed p_Au extends well above 2 V and vanishes at 2.4 V, whereas p_Ag becomes practically zero at around 0.6 V. A small increase in p_Ag around 0.6 V may be a statistical noise and of little significance. This result clearly indicates that Au SACs is more stable than Ag SACs at high biases. It further suggests two other facts on the stability of SACs. First, the marked difference between p_Au and p_Ag tells us that the bias dependence of p of an SAC is not closely related to its electronic structure since Au and Ag SACs show similar electronic structures but have different ps. Second, the critical bias (0.6 V), at which p_Ag vanishes, roughly agrees with those of p_Au (0.8 V and 0.6 V, respectively) [4,7]. This comparison implies that the bias dependence of p_Ag is a ‘normal’ behavior of p, and the wide bias range of p_Au over 2 V is rather exceptional.

FIG. 3: Bias dependence of p_Au and p_Ag. Both p_Au and p_Ag decrease with increasing the bias, but p_Ag vanishes at much lower bias than that of p_Au.

FIG. 4: A typical conductance trace of Au in a high-conductance regime up to 100G₀. Note the nonlinear conductance scale in the plot. The applied bias is 1.6 V, but the real bias voltage across the contact is less than the applied bias and varies with the conductance according to Eq. (2). The conductance trace shows a sudden conductance fluctuation and then abruptly drops to 0. A conductance histogram obtained from the trace is also displayed in a right panel. A peak of the conductance histogram indicates the location of the conductance fluctuation.

B. Conductance fluctuations in the high-conductance regime

Since SACs are formed as a result of contact elongation, its formation probability is likely to depend on necking deformation processes which reduce a macroscopic contact eventually to a SAC. To obtain more information on the evolution of contact deformation, we extended the monitor range of the conductance up to 100G₀ and observed conductance traces in a high-conductance regime. A typical result obtained on Au is displayed in Fig. 4. In this plot, the vertical axis represents V_m in Eq. (1) which was converted to the conductance using Eq. (1).
FIG. 5: Conductance histograms of Au at different biases constructed from 2000 traces. A peak of the conductance profile in each histogram corresponds to the most preferred position of the conductance fluctuation and defines its threshold conductance $G_{th}$. Some histograms at low biases show no peak structures. The peak position, or $G_{th}$, shifts to higher conductance values with increasing the bias. Note again that the bias indicated in each histogram is $V_a$, not the true bias $V_b$ which varies with the conductance according to Eq. (2).

As a result, the conductance scale is quite nonlinear. The conductance trace first decreases smoothly. When the conductance becomes $\sim 20G_0$, it shows a sudden fluctuation and then abruptly drops to 0 due to a sudden contact break. Though this kind of conductance fluctuation does not necessarily appear on every conductance trace, it has a marked tendency to take place not at random position but at certain critical conductance. To quantitatively locate the position of the conductance fluctuation, we constructed a conductance histogram from conductance traces. Such a histogram obtained from the trace of Fig. 4 is also displayed in the figure (this histogram is plotted with the conductance axis upright so that the histogram represents the projection of the trace onto the conductance axis). A peak of the conductance histogram is due to the conductance fluctuation, and hence the peak position indicates the critical conductance for the fluctuation. If the fluctuation takes place at random positions, a summation of many such single histograms would smear out peaks of single histograms and result in no peak structures. We therefore constructed a total histogram from 2000 single histograms (2000 conductance traces) and obtained a result shown in Fig. 5. Since each histogram shows an appreciable background, which grows with increasing the conductance, we first subtracted the background using a power-law fit to it and examined whether the resulting new histogram shows a peak or not. In the case of Au contacts, we could find a broad peak for $V_a > 1.2$ V, which mostly appears as a small shoulder-like structure in the original histogram. At each $V_a$, the position of this peak represents the most preferred onset point of the conductance fluctuation and thus defines a threshold conductance $G_{th}$. We determined $G_{th}$ using a polynomial fit to the peak and the resulting $G_{th}$ is indicated by a red line in each histogram in Fig. 5. The red line shifts to higher conductance side as $V_a$ increases, showing that $G_{th}$ of Au increases with $V_a$.

In the high-conductance regime, $GR_0 >> 1$ in Eq. (2) and $V_a$ much differs from the true bias $V_b$. Therefore, $V_a$ is not a good parameter for discussing the bias dependence of $G_{th}$. Instead, we found that a simple linear relation can be obtained if we plot $G_{th}$ against the contact current $I$ given by Eq. (3). Figure 6 shows $I - G_{th}$ plots for Au contacts. Each point on these plots represents $G_{th}$ obtained at different $V_a$, and the corresponding contact current was calculated from Eq. (3) with substituting $G_{th}$ for $G$. In Fig. 6, data points of Au well lie on a straight line passing through the origin.
a straight line passing through the origin. This line separates two regions on the $I - G_{th}$ plane. Above the $I - G_{th}$ plot, $G > G_{th}$, and the conductance fluctuation makes a contact unstable. On the other hand, below the line, no conductance fluctuations take place and the contact remains stable. Therefore, the straight line in Fig. 6 marks the stable-unstable boundary for Au contacts. The physical meaning of this straight $I - G_{th}$ plot becomes clearer by noting that the conductance in this high-conductance regime can be treated classically and should thus be proportional to a critical current density of the contact, which separate stable and unstable regions for the contact. The existence of the critical current density strongly suggests that the conductance fluctuation is a current effect and signals the onset of certain current-induced contact instability.

We could observe similar conductance fluctuation in Ag contacts as well. The fluctuation was more noticeable than that in Au and appeared almost in all conductance traces. As a result, conductance histograms of Ag showed a well-defined peak, from which we could determine $G_{th}$ and make the $I - G_{th}$ plot of Ag, just as in the case of Au. The Ag $I - G_{th}$ data points again lie on a straight line which passes through the origin, but its slope was merely $\sim 1/3$ of that of Au. This result implies that the critical current density of Ag contacts is three times lower than that of Au. Though the smaller critical current density of Ag is seemingly consistent with the smaller $p_{Ag}$ shown in Fig. 3, our latest measurements yielded different results which suggest higher critical current density for Ag. Probably, the onset of instability in Ag contacts should be much more sensitive to experimental conditions than in Au, and we postpone analyses of $G_{th}$ of Ag until more definite fluctuation data are accumulated.

IV. DISCUSSION

First, we discuss possible mechanisms of the conductance fluctuation, or the current-induced contact instability. Since the conductance fluctuation depends on the current density, natural candidates are Joule heating and electromigration (or their combination). As to Joule heating, the temperature rise in a nanocontact due to electron energy dissipation is still a matter of controversy [1], and no definite results have been established on the contact temperature under high-bias/high-current conditions. On the other hand, electromigration of atoms has been experimentally observed in atom-sized contacts under high biases [8,9] and two-level fluctuations (TLF) in conductance were reported on some metal nanocontacts at low temperatures [8,10,11]. Although these previous observations of TLF suggest that our conductance fluctuations are likely due to electromigration, no detailed analyses can be made at this time since electromigration is a thermally activated process but we lack information on the contact temperature.

Next, we consider how the current fluctuation influences the SAC formation and its probability $p$. When we break a contact, its conductance $G$ decreases with time. Under a constant $V_{a}$, the contact current $I$ also varies with $G$ according to Eq. (3). We plot in Fig. 7 a trajectory $I(G)$ at $V_{a} = 2.0$ V together with the $I - G_{th}$ plot of Fig. 6. Suppose an Au contact at $V_{a} = 2.0$ V. When we break it, the contact current decreases following the $I(G)$ trajectory in Fig. 7 from right to left. The trajectory crosses the $I - G_{th}$ plot around $22G_{0}$ and the contact becomes unstable due to the conductance fluctuation. Most contacts rupture at this point, as exemplified in Fig. 3. However, the occurrence of the conductance fluctuation is also a statistical event and does not have a 100% probability. Some contacts therefore do not suffer instability and some fraction of them can even shrink to a SAC. In fact, $1G_{0}$ SAC plateaus can be observed in Au contacts even at $V_{a} = 2.0$ V (with probability $p_{Au}$ given in Fig. 3). This leads to an idea that SACs under high biases are essentially formed as metastable states, and $p$ represents a formation probability of such metastable states. Then it is natural to consider that the magnitude of $p$ perhaps depends on how large driving force for instability a contact experiences during the SAC formation. As discussed in the previous section, the conductance fluctuation or the contact instability is probably a current-induced effect. Then, the driving force acting on our contact should be measured by an excess current beyond the $I - G_{th}$ plot, as depicted in Fig. 7. Higher the excess current, higher the chance of contact rupture and lower the probability of SAC formation. It is worth noting that this new interpretation of $p$ can naturally account for the suppression of $p_{Au}$ with the bias. With increasing $V_{a}$, the $I(G)$ trajectory in Fig. 7 shifts upward and crosses the $I - G_{th}$ plot at higher current. This generates larger excess current, enhances the driving force for instability, and reduces the SAC formation. Quantitatively, however, we are yet unable to reproduce the $p_{Au} - V_{a}$ curve in Fig. 3 since we do not know how the reduction of $p$ is related to the ex-
cess current. For Ag contacts, it is quite likely that a similar current-induced instability is also responsible for the suppression of $p_{Ag}$. Explanations as to why $p_{Ag}$ is smaller than $p_{Au}$ and vanishes at lower bias will, however, be possible only after carrying out further measurements and establishing the $I - G_{th}$ plot, or the critical current density, of Ag contacts.

V. CONCLUSION

We have studied the high bias conductance of Ag nanocontacts and compared the SAC formation probability of Ag with that of Au. We found that both $p_{Au}$ and $p_{Ag}$ decrease with increasing the bias, but $p_{Ag}$ vanishes at 0.6 V, well below the critical bias 2.3 V of $p_{Au}$. The formation probability thus much differs for Au and Ag SACs though they show the same $I_G$ conductance and have similar electronic structures. These properties of SACs are thus not a determining factor for $p$.

We newly observed the conductance fluctuation in the high-conductance regime and showed that the fluctuation takes place when the contact current density exceeds the critical current density. We consider that this conductance fluctuation indicates the onset of certain current-induced contact instability, perhaps due to electromigration, but many details are still unclarified. The observation of the conductance fluctuation leads us to a new idea that SACs are formed as metastable states against the current-induced contact instability and its formation probability depends on the magnitude of the excess current flowing through the contact. This interpretation of the SAC formation qualitatively accounts for the suppression of $p$ at high biases.

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