Current-induced Instability in Ag and Cu Nanocontacts

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We studied high-bias conductance of Ag and Cu breaking nanocontacts and found that their transient conductance traces often exhibit a sudden conductance fluctuation and a subsequent rapid contact break, the same contact instability as we previously observed on Au nanocontacts [A. Fujii et al., e-J. Surf. Sci. Nanotech. 2, 125 (2004)]. As in the case of Au, the conductance fluctuation tends to take place at a preferred threshold conductance $G_{th}$, which is found to increase in proportion to the contact current $I$. The slope of this straight $I-G_{th}$ plot represents the critical current density $j_c$ for the onset of the conductance fluctuation. Our experimental results show that $I-G_{th}$ plots of Ag and Cu are nearly the same as that of Au, suggesting similar $j_c$ values for three noble metal nanocontacts. [DOI: 10.1380/ejssnt.2004.155]

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I. INTRODUCTION

In our previous paper [1] on breaking Au nanocontacts under high biases, we reported a characteristic conductance fluctuation, followed by a rapid contact break, which often appears in a transient conductance trace when the conductance reaches a certain threshold value. An interesting feature of this contact instability is that the distribution of the threshold conductance is not random but shows a peak, the position of which represents the statistically preferred threshold conductance $G_{th}$. We found that $G_{th}$ is linearly proportional to the contact current $I$ for $G > 10G_0$ ($G_0 = 2e^2/h$ is the quantum unit of conductance). This linear $I-G_{th}$ plot divides the $I-G$ plane into two regions, stable and unstable regions, and a contact becomes unstable against the current-induced instability when the contact current exceeds the critical $I-G_{th}$ line. If we assume that the conductance higher than $10G_0$ linearly depends on the contact area, then the slope of the $I-G_{th}$ plot would represent the critical current density $j_c$ for the onset of the conductance fluctuation. Such instability phenomena in nanocontacts under extremely high current densities are not only of academic interest but also of practical importance in their relations to the reliability of interconnects in nanoelectronics devices. To get further insight into the conductance fluctuation and the contact instability, we extended our experiment to other metals and investigated whether similar conductance fluctuations can be observed in their nanocontacts. We present in this paper our experimental results on Ag and Cu nanocontacts, and will demonstrate that the same contact instability appears in these metals as well.

II. EXPERIMENTAL

Details of measurements have been described in Ref. 1. In short, nanocontacts were formed by repeatedly breaking a junction between a metal wire and a metal plate, employing a piezo actuator. Transient conductance was measured at every contact break by monitoring a voltage drop $V_m$ across a current-sensing resistor $R_0$ (1 kΩ) connected in series with the contact. The conductance $G$ at an applied bias $V_a$ can be obtained from $V_m$ through a following relation,

$$G = \frac{V_m}{R_0(V_a - V_m)}, \quad (1)$$

while the contact current $I$ varies with the conductance as,

$$I = \frac{GV_a}{1+GR_0}. \quad (2)$$

We recorded transient conductance traces for $0 \leq G \leq 100G_0$. We used 99.999 % Ag and Cu samples and carried out all measurements at room temperature in UHV with a base pressure of $\sim 2 \times 10^{-8}$ Pa.

III. RESULTS AND DISCUSSION

We show in Figs. 1 (a) and (b) typical conductance traces of Ag and Cu nanocontacts obtained at $V_a = 2.0$ V, respectively. Note that the conductance scale in both figures is quite nonlinear. This happens because traces in Figs. 1 (a) and (b) are actually $V_m - t$ plots, and we changed their vertical scale from $V_m$ to $G$ using a nonlinear $G - V_m$ relation (eq.(1)). In both traces, the conductance shows a sudden fluctuation and abruptly drops to

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zero. This behavior is just the same as we observed previously on conductance traces of Au [1], and clearly demonstrates that the conductance fluctuation and the current-induced instability also take place in Ag and Cu nanocontacts Comparing conductance fluctuations in Figs. 1 (a) and (b), their amplitudes are more or less similar but the fluctuation in Ag clearly holds on longer than that in Cu (and that in Au as well) and hence appears more noticeable. This tendency has been observed in other conductance traces. We have at this time no adequate explanation why Ag nanocontacts exhibit longer conductance fluctuations.

Following the data analysis employed for Au contacts [1], we constructed conductance histograms for Ag and Cu contacts to determine the threshold conductance \( G_{th} \) at which the conductance fluctuation sets in. The conductance histogram helps us find out \( G_{th} \) because, when the conductance fluctuation appears in one trace, data points recorded during the fluctuation accumulate in a narrow conductance window and form a peak in a conductance histogram of that single trace. This peak would not be smeared out even after adding up histograms of many traces unless the fluctuation takes place at random places. From the position of such a peak, we can know the most preferred \( G_{th} \). Conductance histograms constructed from 2000 traces at different \( V_a \) are summarized in Figs. 2 (a) and (b) for Ag and Cu contacts, respectively.

Each histogram displays a smooth distribution which grows with the conductance. This distribution reflects the fact that the conductance trace first decreases slowly and hence leaves more data points in the high conductance regime. Superposed on this background distribution is a broad peak due to the conductance fluctuation, as we can see in the histogram of Ag at 1.6 V. In Ag histograms, this peak can be well identified for \( V_a \geq 1.2 \) V. On the other hand, in Cu histograms, the peak structure is less evident. This is because conductance fluctuations in Cu nanocontacts are rather short-lived as depicted in Fig. 1 (b), and the number of data points during the fluctuation is too small to produce a distinct peak in a conductance histogram. To find out a peak position of such a small peak, we first approximated the smooth background distribution by a power-law and subtracted it from the histogram. Then we made a polynomial fit to the resulting histogram and obtained the peak position. An example of this procedure is demonstrated in Fig. 3 for the Cu histogram at 1.6 V. A red curve in Fig. 3 (a) displays a power-law fit to the background for \( G > 50G_0 \), which clearly reveals the existence of an additional contribution due to the conductance fluctuation around \((10 - 40)G_0\). The background-subtracted histogram shown in Fig. 3 (b)) now exhibits a well-defined peak structure. We can see from this example that the peak identification can be carried out unambiguously even for seemingly featureless histograms in Fig. 2 (b). The peak position determined in this way is indicated by a red vertical line in each histogram Figs. 2. As mentioned before, each peak position represents the statistically averaged threshold conductance \( G_{th} \) for the onset of the conductance fluctuation. For low biases \( V_a < 1.2 \) V, we could observe the conductance fluctuation less frequently and found no substantial peaks in both Ag and Cu histograms. The conductance fluctuation is thus a high-bias/high-current instability.

In Sec. I, we mentioned that the \( G_{th} \) of Au nanocontacts increases with \( V_a \) and obeys a linear relation \( G_{th} \propto I \) with respect to the contact current \( I \) [1]. The same behavior can also be observed for \( G_{th,S} \) of Ag and Cu contacts. In Fig. 2 (a), for example, the \( G_{th,Ag} \) first appears at \( V_a = 1.2 \) V and shifts to higher conductance values as \( V_a \) increases. Values of \( G_{th,Ag} \) obtained at different \( V_a \) are found to lie on a straight line as plotted in Fig. 4. The same result holds for \( G_{th,Cu} \) equally as well. In Fig. 4, red and blue dashed lines are the least square fit to \( I - G_{th,Ag} \) and \( I - G_{th,Cu} \) plots, respectively. For comparison, we also plotted in the figure the \( I - G_{th,Ag} + \) data (empty circles) obtained in our previous experiment [1].

It can be seen in Fig. 4 that three \( I - G_{th} \) plots are not much different and show similar slopes, though the \( I - G_{th,Ag} \) plot is a little steeper than other plots. This result clearly suggests that for \( G > 10G_0 \), three noble metal
nanocontacts have nearly identical $j_c$s. We can estimate the magnitude of $j_c$ by assuming the following Sharvin conductance (for a circular constriction) [2] which relates $G$ to the contact area $S$ through,

$$\frac{G}{G_0} = \frac{k_F^2 S}{4\pi}$$

where $k_F$ is the Fermi wavelength. Then, three $I - G_{th}$ plots in Fig. 4 yield $j_c \sim (6 - 8) \times 10^{10}$ A/cm$^2$, which is orders of magnitude higher than the current density $\sim 10^6$ A/cm$^2$ at which appreciable electromigration takes place in metal interconnects [3]. Considering this extremely high current density and the observed linear dependence of $G_{th}$ on $I$, the conductance fluctuation, or the contact instability, is probably caused by electromigration and/or local melting (we may need not distinguish these two phenomena since massive electromigration should not be so much different from local melting). If $j_c$ represents the critical current density for such a disruptive electromigration, similar $j_c$ values for Au, Ag, and Cu nanocontacts would not be unexpected since these metals have similar activation energies of electromigration. This coincidence happens because the activation energy of electromigration empirically scales with the melting point [4], which differs less than 10% for Au, Ag and Cu. The observed agreement among $j_c$ values of these noble metals is thus consistent with electromigration.

It is, however, not yet well understood how the electromigration causes the conductance fluctuation. In Fig. 1, we can see that the conductance fluctuation takes place in the middle of a rapid conductance drop and apparently slows down the decrement. This implies that a fast necking deformation of the contact is temporarily suppressed during the conductance fluctuation. We can therefore consider a following scenario: when the current density reaches $j_c$, a massive electromigration takes place in a contact and momentarily recovers the contact necking. Since this contact healing increases the contact area and decreases the current density, the electromigration should halt spontaneously. Then, the necking deformation should proceed again to increase the current density. The observed conductance fluctuation is likely to represent such deformation-recovery cycles. This scenario is physically sound but indeed speculative, and needs further experiments for its justification. Also, the prolonged fluctuations in Ag nanocontacts, mentioned before, remain unexplained. For even smaller contacts consisting of one or a couple of atoms or molecules, there have been a number of recent experimental [5–10] and theor-
FIG. 3: A smooth background in the Cu histogram at $V_a = 1.6$ V can be approximated by a power-law fit (a red curve in (a)), and the background-subtracted histogram shown in (b) reveals a well-defined peak structure.

FIG. 4: $I - G_{th}$ plots for Ag, Cu, and Au nanocontacts. Red and blue dashed lines are the least square fit to $I - G_{th,Ag}$ and $I - G_{th,Cu}$ plots, respectively. Three $I - G_{th}$ plots are not much different from each other.

As we discussed in our previous paper [1], a probability $p$ of the formation of a single atom contact in a breaking contact critically depends on the $I - G_{th}$ plot. This dependence arises from the fact that, under high biases, an $I - G$ trajectory of a breaking contact, described by eq.(2), crosses the $j_c$ line and tracks down through an unstable area in the $I - G$ plane to reach the single-atom contact state. As a result, higher $j_c$ expands stable area for the trajectory and leads to higher $p$. According to this argument, the observed similar $j_c$ values of Au, Ag, and Cu nanocontacts would predict similar $p$s for these metals. However, $p_{Ag}$ is actually much higher than $p_{Au}$ and $p_{Cu}$ [1]. The $j_c$-related instability is thus not sufficient to explain the element dependence of $p$, and an additional instability in Ag and Cu nanocontacts should thus be considered to account for low $p_{Ag}$ and $p_{Cu}$. Such an instability will be discussed in a separate paper [17].

Finally, we add to note that the $I - G_{th,Ag}$ plot of Ag is more sensitive to contamination than those of other two metals and often exhibits a smaller slope, or lower $j_c$, when contacts are not clean. We speculate that this might be due to sulfur contamination. Since $Ag_2S$ is a superionic conductor and experimentally verified to exhibit high electromigration mobility even in atom-sized contacts [18], it is quite likely that the sulfur contamination leads to the $Ag_2S$ formation and makes the contact unstable at lower current density than the $j_c$ of clean Ag contacts.

IV. CONCLUSION

As an extension of our previous work on Au nanocontacts, we measured the transient conductance of breaking Ag and Cu nanocontacts in high-bias/high-current regime and found that the same conductance fluctuation as previously observed in Au takes place in Ag and Cu nanocontacts as well. This conductance fluctuation appears not at random but at a statistically preferred threshold conductance $G_{th}$ which linearly increases with the contact current $I$. By comparing $I - G_{th}$ plots of Au, Ag, and Cu, we showed that these plots have nearly the same slope.
and hence indicate similar values for the critical current density $j_c$ for the conductance fluctuation. The observed agreement among $j_c$ values of three noble metal nanocontacts is consistent with the contact instability due to electromigration. However, this instability alone is not sufficient to account for different formation probabilities of single-atom contacts for Au, Ag, and Cu.

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