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Mechanism of electromigration and spontaneous formation of single electron transistors at ultrasmall copper nano-junctions

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1. Introduction

Electromigration (EM) is a gradual migration process of metal atoms induced by high-density current, which is the main origin of failures in VLSI interconnects. Over the last several years, EM has been attracting attention from different viewpoints; that is, EM is useful in fabricating atomic-scale gaps for interfacing single molecules with macroscopic circuitry ("electrical break junction (EBJ)" process)[1]. Using the EBJ process, Park et al. successfully fabricated nanogap electrodes and captured single molecules, demonstrating that single molecular transistors operate as single electron transistors (SETs) [2].

However, many groups reported that SET behaviors were observed after EBJ even without intentionally introducing molecules and attributed the observed effect to spontaneous formation of metal clusters between the nanogaps after EBJ process [3,4], which makes it very difficult to identify electron transport through single molecules. Therefore, it is important to understand the underlying physics of EM.

So far, EM in bulk metal wires has been explained in terms of two mechanisms. One is "electron wind force" and the other is local Joule heating. These mechanisms seem to be responsible for the formation of nanogaps to some extent. Strachan et al. have succeeded in reproducible fabrication of nanogaps by tuning the applied voltage in such a way that the power dissipation at the nanojunction is kept constant [5]. However, it is also known that significant deviation from the constant power dissipation model is observed when the junction waist becomes as small as several tens of metal atoms [6].

In this work, we have investigated elementary process of EM in ultrasmall Cu junctions. We show that atom migration in such junctions is promoted by microscopic kinetic energy transfer from single conduction electrons to single Cu atoms [7]. We also discuss spontaneous formation of metal clusters during the EBJ process.

2. Experimental

The samples used in the present study were weak Cu nanojunctions fabricated on the surfaces of either oxidized, heavily p-type doped Si substrates or semi-insulating GaAs substrates. Cu nanojunctions were fabricated by electron beam lithography using double layer resist (PMMA/MMA) and subsequent shadow evaporation. At the first step of the shadow evaporation, a 3 nm-thick Ti layer was deposited at normal incidence as an adhesion layer and, then, 12nm and 18nm thick Cu layers were deposited at incidence angles of $+11^{o}$ and -11°, respectively, to form a small overlapping area, as shown in the inset of Fig.1(a). By doing so, the weakly overlapping region was free from the Ti adhesion layer. The waist of the fabricated nanojunctions was typically 120nm. Finally, 10nm Cu layer was deposited at normal incidence to ensure a reliable bonding between copper nanowires and gold bonding pads. All the experiments were performed in liquid He.

3. Results and Discussion

We applied the EBJ process to Cu nanojunctions and fabricated atomic scale gaps. We increased the bias voltage until small reduction in the sample conductance was detected and, then, relaxed the bias voltage by 100mV, and did the same process repeatedly. Using this feedback-controlled EBJ technique, atoms can gradually be removed without catastrophic breakage. By this technique we have successfully fabricated atomic scale gaps. Fig.1(a) shows a typical conductance-voltage (G-V) record during the entire FC-EBJ process. As seen in Fig. 1(a), the junction conductance was gradually reduced below G_0, where $G_0 \equiv 2e^2/h$.

During the EM, the conductance gradually decreases from its initial value G_S. Since this conductance decrease can be attributed solely to the development in EM, we defined the local conductance of the junction, G_J, by $1/G_J = 1/G-1/G_S$, and the junction voltage, V_J, by $V_J = I/G_J$. Using these definitions, the G-V data shown in Fig.1(a) can be replotted into the time evolutions of G_J and V_J, as shown in Fig.1 (b) and (c). By comparing Figs.1(b) and 1(c), we have found that the mode of EM changes as EBJ proceeds. At the initial stage denoted as phase I in Fig. 1(b), where G_J is as large as a few hundreds of G_0, G_J exhibits repetitive sharp decreases and incomplete recoveries, synchronizing to stress and relax voltage control. However, as G_J decreases down to several tens of G_0, G_J starts showing quantized conductance plateaus and successive stepwise drops by G_0, as seen in Fig.1(c). To gain more insight into elementary processes, we paid attention to the critical value of the junction voltage, V_c, which we defined as the voltage at which significant reduction in G_J starts to appear. In Fig. 2, we plotted V_c as a function of R_J. As seen in the figure, V_c increases with increasing R_J. When R_J is small (phase I), the relationship between R_J and V_c can be well fitted by the conventional Joule heating model, $R_J=V_c^2/P^*$, where P^* is a constant and has been interpreted as a critical power dissipation to initiate EM at the junction [6]. As the junction waist becomes as narrow as several tens of atoms

(phase II), however, the R_J-V_c relationship significantly deviates from the constant power line, suggesting that a non-thermal mechanism sets in. This is further supported by the fact that the electron transport when G_J < several tens of G_0 is ballistic. In this regime, as seen in the Fig.2, critical voltage for various samples gradually approached ~0.8 V. A remarkable finding is that 0.8 eV is very close to the activation energy for the mean time to failure (MTF) determined empirically from the reliability data for Cu wires on SiO_2, 0.79 ± 0.02 eV [8]. This fact clearly indicates that the elementary process of EM is not Joule heating nor electron wind force, but the self-diffusion of a metal atom driven by microscopic kinetic energy transfer from single conduction electrons to single atoms.

In the inset of Fig.1(a), an SEM image after EBJ process was shown. Almost all samples show deformation of the electrode after EBJ process. This is due to local melting during phase I of the FC-EM process. We sometimes experienced sudden large change in G_J in phase I during Fc-EBJ and G-V trace exhibited reduction down to $G < 10G_0$ and subsequent gradual decrease to $G < G_0$, as seen in Fig. 3(a). After this EBJ process, the sample showed Coulomb blockade effect. Similar behavior was observed also for Au and Pt junctions. Therefore, sudden resistance change in phase I results mostly from spontaneous formation of metal clusters in the gap. This indicates local Joule heating and melting has a crucial role for cluster formation in the gap.

The cluster was confirmed by 3-terminal electrical measurements. The substrates were used as gate electrodes. As shown in Fig.3(b), oscillatory behavior was clearly observed in the conductance spectrum, which is characteristic of single electron tunneling and indicates that the nanojunction works as a SET, even though molecules were not intentionally introduced. The observed Coulomb stability diagram suggests that there are a few Coulomb islands in the nanogap region. This is supported by SEM images (not shown); they show several metallic dots whose diameter is less than 10nm in the gap region.

4. Conclusion

We have investigated the elementary process of EM in copper nanojunctions. When the junction conductance is less than $50G_0$, we observed successive step-like decreases in the junction conductance by one or integer multiples of the quantum conductance, which took place only when the junction voltage exceeded certain critical voltages. It is found that the critical voltage has a good agreement with the activation energy of the MTF of copper wires. This fact indicates that the elementary process of EM in the ballistic regime is the atom diffusion driven by microscopic kinetic energy transfer from single conduction electrons to single metal atoms, as in the case for gold nanojunctions [7]. In addition, we have found that local Joule heating and subsequent melting has a crucial role for spontaneous formation of metal cluster in the nanogap region. This knowledge of EM physics is useful for reproducible fabrication of single molecular transistors.

References

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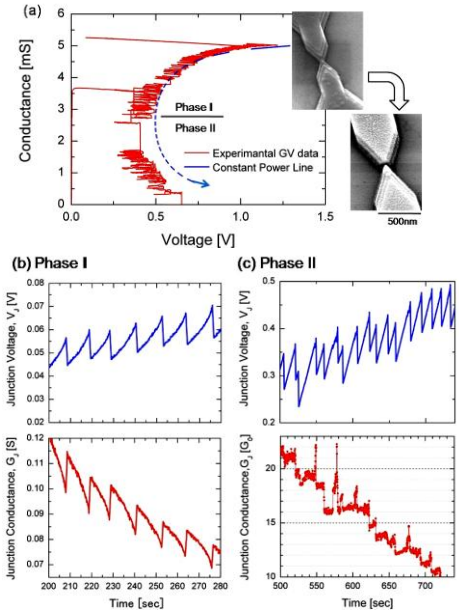

Fig.1 (a) Conductance-Voltage record during FC-EM process. The inset shows the SEM images of before and after EBJ. Time evolution of the junction conductance G_J and the junction voltage V_J, when G_J~$150G_0$ (phase I) (b) and G_J=$20G_0$ (phase II) (c).

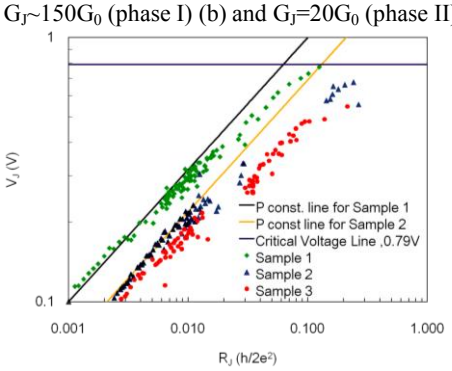

Fig.2 The critical voltage V_c is plotted as a function of the junction resistance R_j during the entire EBJ process for three samples.

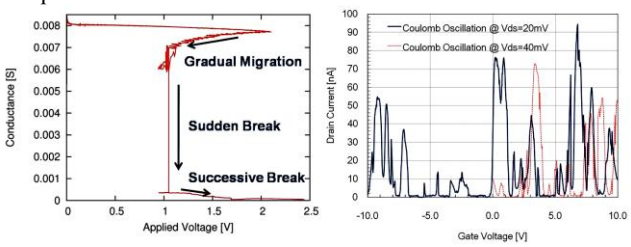

Fig.3 (a) FC-EM process resulted in the spontaneous formation of metal clusters in the nanogap, (b) A conductance spectrum of the sample measured at drain voltage = 20mV and 40mV. A Coulomb oscillation-like feature was observed.