Fabrication and Evaluation of 20-nm-Wide Intercalated Multi-Layer Graphene Interconnects and 3D Interconnects Composed of Graphene and Vertically Aligned CNTs

Daiyu Kondo^a, Haruhisa Nakano^a, Bo Zhou^a, Ichiro Kubota^a, Kenjiro Hayashi^a, Katsunori Yagi^a Makoto Takahashi^a, Motonobu Sato^a, Shintaro Sato^a and Naoki Yokoyama^a

^a Collaborative Research Team Green Nanoelectronics Center, AIST, 16-1 Onogawa, Tsukuba 305-8569, Japan, kondo.daiyu@aist.go.jp, shintaro.sato@aist.go.jp

Introduction: Nano-carbon materials including graphene [1] are one of promising electronic materials for future LSIs due to their excellent physical properties. Especially their tolerance to a high current density might make nano-carbon materials replace cupper (Cu) as future interconnect materials, since the reliability of Cu interconnects decreases with their line width. As for resistivity, however, nano-carbon materials have generally shown a resistivity higher than that of Cu. For instance, high-quality graphene peeled off from HOPG has a resistivity of ~40 $\mu\Omega$ cm, an order of magnitude higher than Cu. Recently, we reported that intercalated MLG interconnects with a width of 4 μ m formed by chemical vapor deposition (CVD) had a resistivity of the same order as Cu [2]. However, it is not clear whether MLG interconnects with a line width below 100 nm can also exhibit such a low resistance.

In this study, we fabricated intercalated MLG interconnects and obtained a resistivity as low as Cu. Furthermore, intercalated MLG interconnects with a width of 20 nm were fabricated and evaluated. We also performed reliability tests at 250°C, showing that the MLG interconnects with a width of 20 nm and a thickness of ~3.7 nm (11 graphene layers) had high-current reliability superior to Cu.

Results and discussion: The fabrication process for MLG interconnects is described shown in Fig. 1. A schematic diagram of a MLG interconnect with four terminal electrodes is shown in Fig. 2. Shown in the inset is a SEM image of a MLG interconnect with electrodes. MLG grown on epitaxial cobalt film on a sapphire substrate indicated a resistivity as low as ~50 $\mu\Omega$ cm [2]. A reliability test showed that the MLG was not damaged at a current of 1×10^7 A/cm² after 219 hours at 250°C, demonstrating reliability higher than Cu [3]. After intercalation of FeCl₃ into MLG, which was confirmed by Raman spectroscopy, the cumulative probability distribution of sheet resistance of intercalated MLG in Fig. 3 indicates that the intercalation (R₁) to the original resistance (R₀) was estimated to be 0.046. From the four-terminal I-V measurements, the lowest resistivity was found to be 4.1 $\mu\Omega$ cm for an interconnect with a length of 6 μ m and a width of 4 μ m. This value is of the same order as that of bulk Cu.

After the MLG interconnects with widths wider than 1 µm were evaluated, they were patterned by EB lithography and etching to form 20-nm-wide interconnects. The widths of narrow MLG interconnects were confirmed to be ~20 nm by SEM (Fig. 4 (a and b)). The resistivity of a 20-nm-wide MLG interconnect with a length of 4 μ m was measured to be 25 μ Ωcm, while that of the original 20- μ m-wide interconnect before EB-patterning had been 16 $\mu\Omega$ cm (the resistivity was relatively high due to a shorter intercalation period of 12 hours). The resistivity did not increase so much by narrowing down the interconnect to 20 nm. In fact, four MLG interconnects with a length of 4 µm exhibited almost no change in resistivity after patterning to 20 nm shown in Fig. 5. This is in good contrast with the case of Cu, whose resistivity increases with decreasing width below 100 nm [4]. On the other hand, the resistivity of MLG interconnects longer than 4 µm increased after the 20-nm patterning. This might be related to the grain size of CVD graphene. A reliability test of a 20-nm-wide MLG interconnect with a thickness of ~3.7 nm (11 graphene layers) at a current of 1×10^7 A/cm² and a temperature of 250°C showed that the graphene interconnect did not break over 110 hours, exhibiting higher reliability than 160-nm-wide Cu interconnect (Fig. 6). These results showed that intercalated MLG has become a really promising candidate for future scaled LSI interconnects. We also fabricated 3D interconnect structures consisting of horizontal MLG interconnects and CNT vias, although the results are not shown here due to the space limitation.

Conclusion: We fabricated 20-nm-wide intercalated MLG interconnects and demonstrated their higher reliability than Cu. The intercalated MLG showed a resistivity of 4.1 $\mu\Omega$ cm and the resistivity did not increase by narrowing down the interconnect to 20 nm.

Acknowledgements: This work was supported by JSP through the "FIRST Program," initiated by CSTP.

References:

[1] K.S. Novoselov, A.K. Geim, S.V. Morozov, D. Jiang, Y. Zhang, S.V. Dubonos, et al., " Electric Field Effect in Atomically Thin Carbon Films," Science, vol. 306, no. 5696, pp. 666-669, October 2004.

[2] D. Kondo, H. Nakano, B. Zhou, I. Kubota, K. Hayashi, K. Yagi, et al., "Intercalated multi-layer graphene grown by CVD for LSI interconnects," IEEE International Interconnect Technology Conference 2013, pp 190-192.

[3] M. Sato, M. Takahashi, H. Nakano, T. Murakami, A. Kawabata, M. Nihei, et al., " Multi-layer Graphene Wire Grown by Annealing Sputtered Amorphous Carbon," IEEE International Interconnect Technology Conference 2012, pp. 1-3.

[4] A. Naeemi, J.D. Meindl, "Compact Physics-Based Circuit Models for Graphene Nanoribbon Interconnects," IEEE Transactions on electron devices, vol. 56, no. 9, pp. 1822-1833, September 2009.

- Catalyst deposition at 500°C
- (Cobalt film=200 nm on a 3-inch sapphire substrate) Graphene growth by thermal CVD method
- (At 1000°C with CH₄ diluted by Ar and H₂)
- Graphene transfer (PMMA)
- Isolation of graphene
- (Photo lithography(TSMR), RIE; O₂, 100 W)
- Metal electrodes formation
- 4-terminal IV measurement
- Intercalation of FeCl₃ (At 310°C for 12-38 hours)
- Metal electrodes formation
- (Photo lithography (TSMR/PMGI), Ti/Au=5/300 nm)
- 4-terminal IV measurement
- EB Lithography (HSQ)

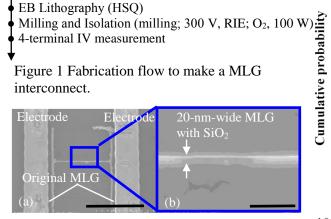


Figure 4 (a) SEM image of 20-nm-wide $\vec{\epsilon}$ intercalated MLG interconnect with a length of $\mathbf{\vec{z}}_{-1}$ 6 µm. The scale bar is 2 µm. (b) Enlarged SEM image of the interconnect. Isolated 20-nm-wide graphene with 70-nm-thick SiO₂ (HSO) used for EB mask is clearly observed. The scale bar is 200 nm.

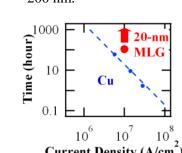


Figure 6 Time to failure for the 20-nm-wide intercalated MLG with a length of 4 µm and a thickness of ~3.7 nm (11 graphene layers) is shown as a red colored circle. The MLG at a current of 1×10^7 A/cm² at 250°C did not break over 110 hours, showing that the graphene interconnect has , higher reliability than Cu with a width of 160 nm,

10

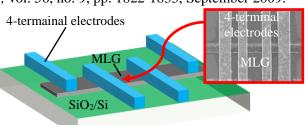
0.1

н

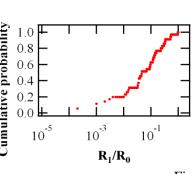
2

4

6



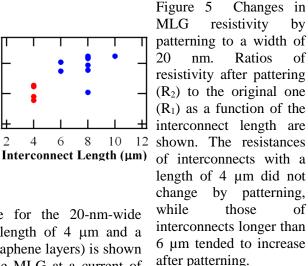
(Photo lithography (TSMR/PMGI), Ti/Au=5/300 nm) Figure 2 Schematic diagram of a MLG interconnect. The MLG was evaluated with four-terminal electrodes which were composed of Ti/Au. A SEM image of the MLG interconnect is shown in the inset.



8

10

Figure 3 The cumulative probability distribution of the ratios of the sheet resistance of intercalated MLG for 38 hours (R_1) to the original (\mathbf{R}_0) .



Current Density (A/cm^{*}) as shown in blue [3].