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Intercalated Multi-layer Graphene Wire and Metal/Multi-layer Graphene Hybrid Wire Obtained by Annealing Sputtered Amorphous Carbon

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Abstract

We have fabricated intercalated multi-layer graphene (i-MLG) wire and cobalt metal/multi-layer graphene (Co/MLG) hybrid wire to decrease the resistivity of multi-layer graphene (MLG) grown by annealing sputtered amorphous carbon. The resistance of i-MLG and Co/MLG wires is reduced by one order and two orders of magnitude to that of MLG wire, respectively. The breakdown tests of the i-MLG and MLG wires indicate that the current tolerance per graphene layer is almost the same, independent of whether the wire is intercalated. On the other hand, the metal/MLG hybrid wire not only exhibits decreased resistivity but also exhibits increased current tolerance.

1. Introduction

Carbon-based materials, such as carbon nanotubes (CNTs) and graphene nanoribbons (GNRs) have been studied as interconnect materials because they have lower resistivity [1], higher thermal conductivity [2], and intrinsically higher current-carrying capacity [3] than Cu. In the past, we reported the resistivity [4] and the current tolerance [5] of multi-layer graphene (MLG) wire fabricated by annealing a sputtered carbon film with a catalyst layer. In this study, we fabricated intercalated multi-layer graphene (i-MLG) wire and cobalt (Co) metal/multi-layer graphene (Co/MLG) hybrid wire to decrease the resistivity of MLG grown by annealing sputtered amorphous carbon.

2. Experimental Materials

We used the magnetron sputtering method to deposit carbon and Co catalyst layers onto a SiO_2/Si substrate at room temperature. The thicknesses of the deposited layers were Co(50nm)/C(30nm) on SiO_2/Si. The film was annealed in an IR furnace in a nitrogen flow atmosphere at a temperature of 800°C for 30 min [4, 5]. The Co/MLG-hybrid wire was obtained by isolating the annealed film. The MLG wire was obtained by isolating the MLG film after removing the Co layer from the annealed Co/MLG film. The i-MLG wire was obtained by intercalating iron chloride ($FeCl_3$) into the isolated MLG wire. The intercalation was performed in a quartz tube with anhydrous $FeCl_3$ powder and the MLG wire at 310℃ for 12 hours. UV lithography was used to produce wires with a width in the range of 1-10μm and a length in the range of 2-400μm. Transfer processes were not used to produce wires in this study.

The nanostructures of the films were analyzed by Raman spectroscopy using an exciting laser wavelength of 532 nm and X-ray diffraction (XRD) using Cu Kα radiation. The thickness of the wires was measured using the Alpha-step. The resistance of the wires was measured by the four-terminal method.

3. Results and Discussion

Figure 1 shows the Raman spectra of MLG and i-MLG. An upwards shift of the G band by 25 cm^{-1} is observed after intercalation (see insert in Fig. 1), as reported in [6]. The amount of shift indicates that the intercalation was successful.

Figure 2 shows the resistance reduction ratios of i-MLG and Co/MLG. The resistances of i-MLG and Co/MLG wires are one order and two orders of magnitude lower than that of MLG wire, respectively, independent of wire width and length. Next, we investigate film thickness to calculate the resistivity of the wires.

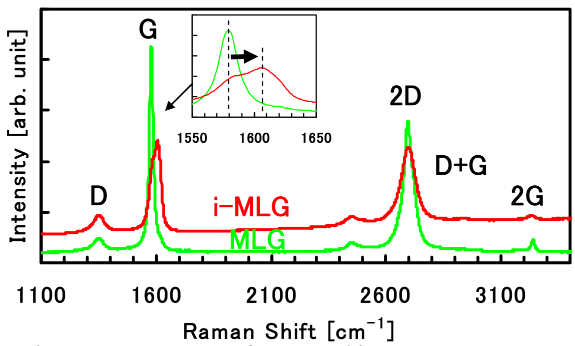

Fig. 1 Raman spectra of MLG and i-MLG

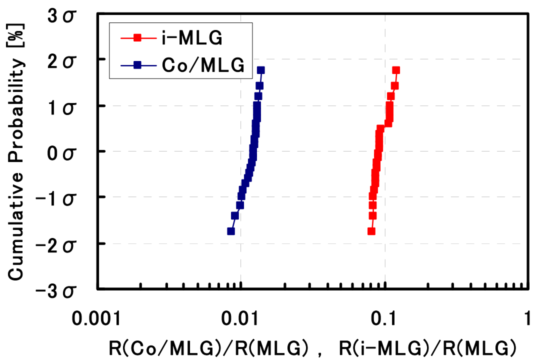

Fig. 2 Resistance reduction ratios of Co/MLG wires and i-MLG wires to MLG wires

Figure 3 shows the thicknesses of Co/MLG, MLG, and i-MLG. The profiles indicate the path of the Alpha-step stylus crossing the wires. The Co/MLG wire has a thickness of approximately 80 nm, the same as the thickness targeted during sputtering deposition. The MLG wire has a thickness of approximately 30 nm after removing the Co layer from the annealed Co(50 nm)/MLG(30 nm). The i-MLG wire has a thickness of 50 nm. Intercalation increases the thickness of the 30 nm MLG wire by 1.6 times. Based on these thicknesses, the MLG, i-MLG and Co/MLG wires have a resistivity of 500 μΩcm [4, 5], 80 μΩcm, and 13 μΩcm, respectively.

Figure 4 shows XRD patterns (θ-2θ). The d-spacing of MLG(002) and i-MLG(002) is 0.338 nm and 0.554 nm, respectively. The distance between graphene sheets after intercalation increased 1.6 times over that before intercalation. This result agrees with the measured thicknesses of the wires and indicates that the number of graphene sheets in both wires is almost the same.

Figure 5 shows the temperature dependence of resistance measured using a low current to prevent Joule heating. The temperature coefficient of MLG is negative while all other materials have positive coefficients. The temperature coefficient of i-MLG becomes between highly oriented pyrolytic graphite (HOPG) and MLG.

Figure 6 shows the results of the breakdown tests performed on the wires. The resistance of the MLG wire decreases as the wire's temperature increases due to Joule heating as a consequence of having a negative temperature coefficient. Conversely, the resistance of the i-MLG and Co/MLG wires increases because of their positive temperature coefficients. The breakdown current density of the Co/MLG wire is higher than that of the MLG wire. On the other hand, the cross-sectional breakdown current density of the i-MLG wire is approximately 1.6 times lower than that of the MLG wire. However, the breakdown current and the number of graphene sheets are actually almost the same in the both wires. Therefore, it can be concluded that the breakdown current density per graphene layer is basically the same and does not depend on whether MLG wires are intercalated or not.

4. Conclusion

We have fabricated i-MLG and Co/MLG wires with resistivity of 80 μΩcm and 13 μΩcm, respectively. Test results indicate that the current tolerance of the graphene sheet in i-MLG does not change by intercalating $FeCl_3$ to MLG, even though its resistivity is significantly reduced. On the other hand, the Co/MLG hybrid wire has both decreased resistivity and increased current tolerance, and has potential uses in interconnects and redistribution layout (RDL) for 3D applications.

Acknowledgements

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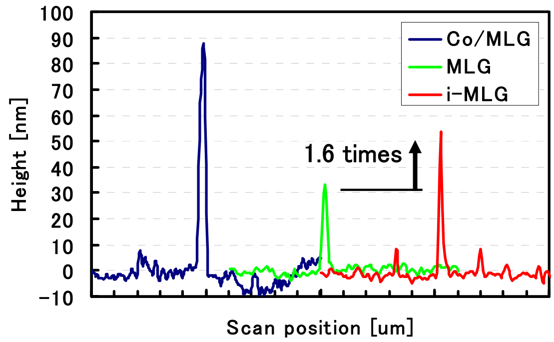

Fig. 3 Thickness of Co/MLG, MLG, and i-MLG wires measured by using the alpha-step

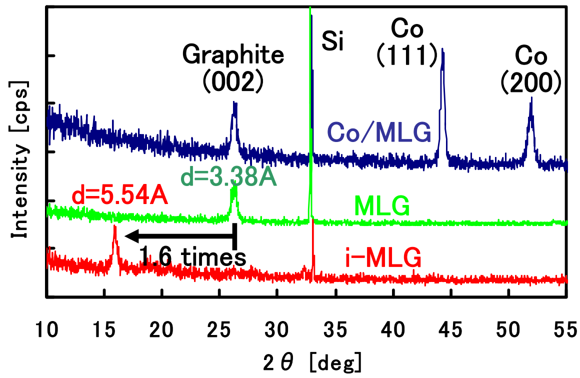

Fig. 4 XRD patterns (θ-2θ) of Co/MLG, MLG, and i-MLG films

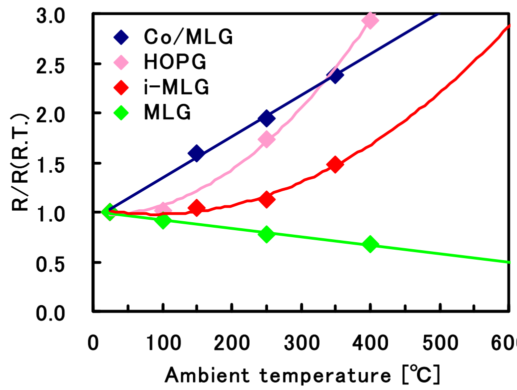

Fig. 5 Temperature dependence of sheet resistance for Co/MLG, MLG, i-MLG, and HOPG

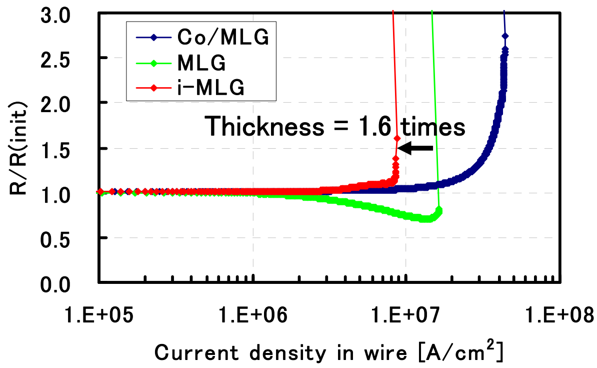

Fig. 6 Resistance changes of wires with increasing current density per cross-sectional area at breakdown tests