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Multi-layer graphene interconnects grown by CVD for future LSI

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Abstract

Multi-layer graphene (MLG) wires were fabricated and evaluated for future LSI interconnects. MLG was grown epitaxially on an epitaxial cobalt film by chemical vapor deposition (CVD). Analyses of the MLG by low energy electron microscopyand four-terminal I-V measurements revealed that the structure and electrical properties of the MLG were comparable with those of a high-quality graphite crystal. It was also found that the MLG wires surpass Cu wires in reliability. The MLG wires were then intercalated with $FeCl_3$. By optimizing the intercalation, the resistivity as low as 4.1 μΩcm was obtained, being almost the same as that of Cu. Our results indicate that intercalated MLG is a promising candidate for future LSI interconnects.

1. Introduction

Two-dimensional (2D) nano materials such as graphene [1, 2] are a candidate for next-generation electronic materials due to their excellent properties. The application of multi-layer graphene (MLG) to interconnects is one of long-awaited technologies in the future LSIs because an ever-increasing current density in LSIs makes Cu interconnects more and more vulnerable to electro-migration.

It has been demonstrated that graphene and carbon nanotubes can sustain a current density higher than the upper limit of Cu [3]. On the other hand, the resistivity of such nanocarbon materials grown by chemical vapor deposition (CVD) is usually worse than that of Cu by more than one order of magnitude [3, 4]. This is really a serious problem in applying nanocarbon materials to LSI interconnects. In this study, we grew high-quality MLG by CVD and fabricated MLG wires whose resistivity was as low as that of Cu for the first time. To achieve this, we employed an epitaxial cobalt (Co) film to grow MLG epitaxially [5]. We have further developed a process to intercalate MLG with $FeCl_3$ to drastically reduce the resistance.

2. Experiment

MLG was synthesized by thermal CVD in a low pressure chamber (Aixtron K.K. Black Magic CRIUS-R) with CH_4 diluted by Ar and H_2 as the source gas. The growth temperature was ~1000℃. As a catalyst, 200-nm cobalt film was deposited by sputtering at 500℃ (ULVAC CS-200). After synthesis, MLG was evaluated with optical microscope, Raman spectroscopy (Horiba Jovan-Yvon; Laser excitation wavelength: 488 nm), scanning electron microscopy (SEM; Hitachi S-4800), X-ray diffraction (XRD; Rigaku Ultima X), electron backscattered diffraction (EBSD; JEOL JSM-6500F), and transmission electron microscopy (TEM; Hitachi H-9000UHR III). The MLG was also analyzed with low energy electron microscopy (SPELEEM; Elmitec LEEM-III) at BL17SU, SPring-8, Japan.

The MLG wires were fabricated with the conventional photo lithography. After synthesis of MLG it was coated with polymenthyl methacrylate (PMMA), and then detached from the substrate by etching the catalyst metal film with ferric chloride ($FeCl_3$). The MLG film was transferred onto a 3-inch Si substrate with a thermal SiO_2 layer. After removal of PMMA, MLG wirings were isolated using reactive ion etching (RIE). The four terminal electrodes were then formed, which were consisted of an Au layer (300 nm) and a Ti layer (10 nm) by the electron beam deposition. Finally, anhydrous $FeCl_3$ was intercalated into MLG at 310℃in the same manner as reported previously [6-8]. The electrical measurement was performed with four-terminal I-V measurements (Keithley 4200-SCS).

3. Results and discussions

Figure 1(a) shows the XRD patterns after CVD synthesis. A sharp peak was observed at 44 degrees which indicates that the film consists of hexagonal close packed (hcp) (0001) Co confirmed with the EBSD image (not shown). A graphite (002) peak was also found at 26.5 degrees, suggesting MLG formation. From a cross-sectional TEM image in Fig. 1(b), MLG synthesis was clearly observed. An optical microscope image of the MLG shown in Fig. 1(c) indicates that there are several regions with different contrasts, implying variations in the number of layers. In addition, the shape of G' band located at ~2700 cm^{-1} in the Raman spectrum shown in Fig. 1(d) is similar to the one observed for highly oriented pyrolytic graphite (HOPG). This feature, along with negligible D-band at ~1350 cm^{-1}, indicates that the synthesized multi-layer graphene is of high quality with an AB-stacked structure.

Figure 2 (a and b) indicate a dark field (DF) LEEM image and a typical μLEED pattern of MLG on Co film.

Field of view (FOV) of DF LEEM image was 20μm. The energy of the electron beam was 48.28eV. The DF image looks almost uniform, indicating that the orientation of MLG is almost the same within this FOV. Furthermore, LEED spots of MLG (not shown) obtained from an area of 20 μm were the same as μLEED spots shown in Fig. 2(b). Although there were variations in the number of layers shown in Fig. 1(c), we found that most of the graphene regions were in the same crystal orientation, matching the underlying Co film.

Four-terminal I-V characteristics and an SEM image of an MLG wire with a length of 6 μm and a width of 4μm are shown in Figs. 3(a) and (b). The resistivity of MLG was estimated to be 56 μΩcm, which is comparable to that of HOPG, indicating that high-quality graphene was obtained. We further evaluated the reliability of MLG wires, and found that the reliability is actually better than Cu (not shown). To improve the resistivity of MLG, intercalation of $FeCl_3$ to MLG was performed. Figures 3(c) and (d) indicate the I-V characteristics, and Raman spectra after intercalation. A shift of G and G' bands by ~30 cm^{-1} to the higher wave number side was observed after intercalation. The amount of shift indicates that the intercalation was successful, although the sample was not probably fully intercalated in this particular case [8]. From the four-terminal I-V characteristics, the resistivity was estimated to be 4.1 μΩcm for wiring with a length of 6 μm and a width of 4 μm. This value is almost the same as that of Cu, and much better than the previously reported values [9]. The cumulative probability distribution of ratios of resistivity after intercalation (R_1) to the original resistivity (R_0) is also shown in Fig. 3(e). The median was estimated to be ~0.046.

4. Conclusions

We have fabricated interconnects using multi-layer graphene (MLG) grown by thermal CVD method on an epitaxial Co film. The MLG exhibited a resistivity as low as that of HOPG. Furthermore, MLG intercalated with $FeCl_3$ was found to be almost the same as that of Cu. With the low resistivity shown experimentally in this study, MLG has now become a really promising candidate for future LSI interconnects.

Acknowledgements

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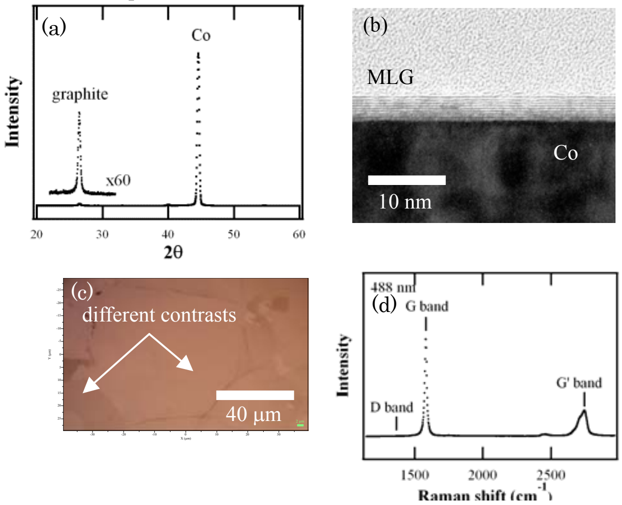

Fig 1. (a) XRD patterns, (b) TEM image, (c) Optical microscope image and (d) Raman spectrum.

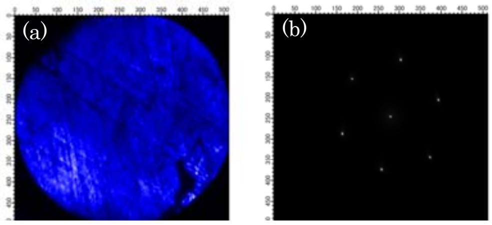

Fig 2. (a) DF image (FOV=20 μm), and (b) micro LEED spot of MLG.

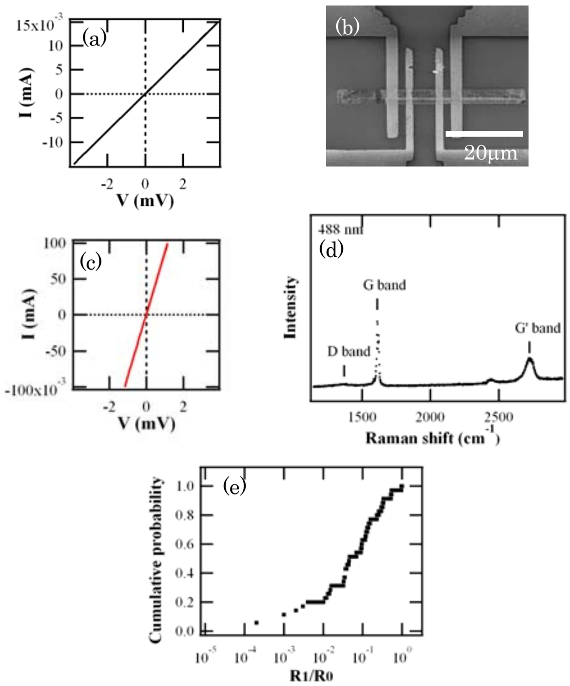

Fig 3. (a) Four-terminal I-V characteristics before intercalation, (b) SEM image of MLG wiring with four terminal electrodes, (c) Four-terminal I-V characteristics after intercalation, (d) Raman spectra after intercalation, and (e) Cumulative probability distribution of the ratios of resistivity (R_1) after intercalation to the original (R_0)