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Transverse Thermoelectric Effect of Graphene Isotopic Heterostructures

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Abstract

Transverse thermoelectric effect requires anisotropic electrical and thermal properties. In this study, we used graphene composed of ^{12}C and ^{13}C to produce isotopic graphene heterostructures with anisotropic properties. Thermoelectric voltage in perpendicular direction to the temperature difference was measured as functions of the interface tilt angle and the aspect ratio of the devices. The results indicate that transverse thermoelectric effect was observed using graphene isotopic heterostructures.

1. Introduction

Graphene is one of the potential candidates for high efficiency thermoelectric conversion elements due to its high power factor. As the thermoelectric performance is positively correlates with power factor and negatively correlates with thermal conductivity, extremely high thermal conductivity prevents graphene from being used for thermoelectric materials with high figure of merit (ZT). To increase ZT values of graphene, the reduction in thermal conductivity of graphene has been investigated using carbon isotopes [1,2] and structural defects [3]. However, ZT of graphene is still less than the value required for practical application. Therefore, we focused on transverse thermoelectric effect, in which thermoelectric voltage appears in the direction perpendicular to the temperature difference. The off-diagonal term of Seebeck tensor in graphene is normally zero because graphene is isotropic materials. In this study, we synthesized graphene heterostructures using isotopic carbon atoms, ^{12}C and ^{13}C, to show transverse thermoelectric effect of graphene. In isotopic graphene heterostructures, the thermal conductivity can be dramatically reduced, leading to the expression of off-diagonal term of Seebeck tensor. As a result, the transverse thermoelectric effect can be expected.

2. Fabrication process

Graphene was synthesized by low-pressure chemical vapor deposition (CVD). Figure 1 shows the fabrication process of isotopic graphene heterostructures. At first, Cu foils treated with HNO_3 were annealed at 1050 °C in the hydrogen flow of 50 sccm for 30 min. Subsequently, graphene composed of ^{12}C and ^{13}C was synthesized with H_2 (150 sccm), $^{12}CH_4$ (0.5 sccm), and $^{13}CH_4$ (0.5 sccm) at 1050 °C for 15 min. After synthesizing graphene composed of 50% ^{13}C and 50% ^{12}C ($^{12+13}$C-graphene), graphene was then photolithographically patterned by O_2 plasma etching. Finally, graphene with only ^{12}C (^{12}C-graphene) was synthesized at the part of the etched region to form isotopic heterostructures. ^{12}C-graphene was grown with 150 sccm of H_2 and 0.4 sccm of $^{12}CH_4$ at 1050 °C for 10min.

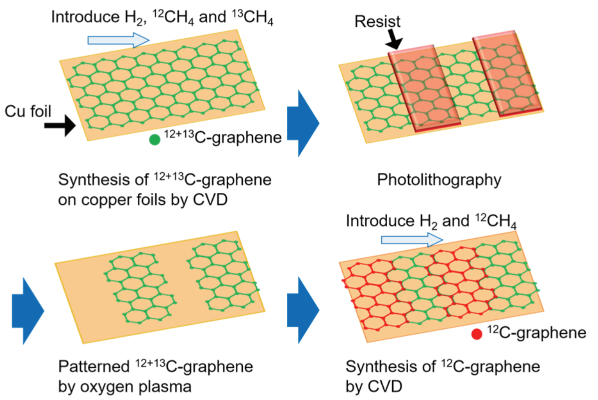

Fig. 1 Fabrication process of graphene isotopic heterostructures.

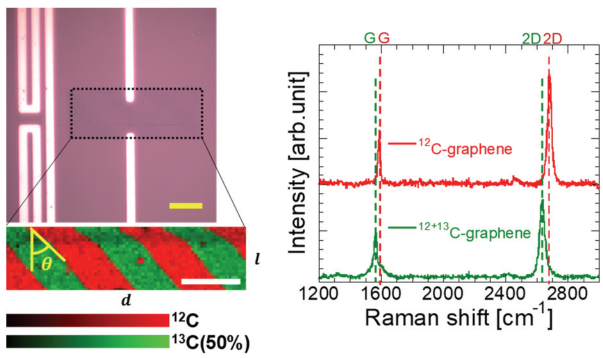

Fig. 2 Raman mapping image and Raman spectra of isotopic graphene heterostructures. Bars in optical microscope image represent 10 μm.

3. Results and Discussion

Figure 2 shows the representable Raman 2D mapping image and Raman spectra of synthesized isotopic graphene heterostructures. The Raman peaks of $^{12+13}$C-graphene appear at lower wavenumbers than that of ^{12}C-graphene because of the mass difference. From Raman spectra, no D band is observed, indicating graphene with almost no defects.

Figure 3 shows the typical transfer characteristics of graphene samples with the channel length and width of 10 μm and 5 μm, respectively. The carrier mobility of each graphene sample was 805 cm^2/Vs (^{12}C-graphene) and 882 cm^2/Vs ($^{12+13}$C-graphene), suggesting that isotopes do not modify the electrical properties of graphene.

Transverse thermoelectric voltage appeared in the perpendicular direction to the temperature difference is expressed as

$$\left|\Delta V\right| = \left|\frac{\Delta T}{2}\frac{l}{d}\left(S_{//} - S_{\perp}\right)\sin 2\theta\right|, \quad (1)$$

where ΔT, l/d, θ, $S_{//}$, and $S_{\perp}$ are the temperature difference, aspect ratio of the device, tilt angle of the isotopic interface, and Seebeck coefficient of graphene heterostructures in the parallel and perpendicular directions to the interface, respectively. The transverse thermoelectric voltage was measured with lock-in amplifier by applying an alternating voltage to the heater to induce the temperature difference across the graphene device (Fig. 4(a)).

Figure 4(b) shows the transverse thermoelectric voltage of graphene heterostructures as a function of tilt angle. The maximum value of the measured transverse thermovoltage is approximately 12.0 μV at the tilt angle of 45°. The trend of transverse thermoelectric voltage fits well to Eq. 1. The aspect ratio dependence of the thermovoltage was also measured, which also fits well to Eq. 1 (Fig. 4(c)). These results suggest that the measured voltage is the transverse thermoelectric voltage as a result of the anisotropicity of graphene induced by the isotopic heterointerfaces, which is useful for high efficiency graphene thermoelectric devices.

3. Conclusions

In this study, we investigated the transverse thermoelectric effect using graphene isotopic heterostructures. The transverse thermovoltage was observed in the perpendicular direction to the temperature difference. We also confirmed the tilt angle and aspect ratio dependence of the voltage, which results from the anisotropicity of graphene heterostructures. The transverse thermoelectric effect may be useful as a novel root for enhancing the thermoelectric performance of graphene devices.

Acknowledgements

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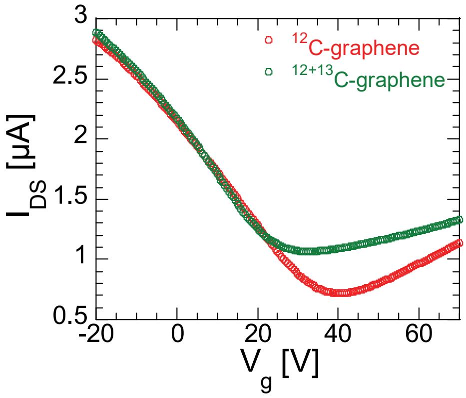

Fig. 3 Transfer characteristics of ^{12}C-graphene and $^{12+13}$C-graphene.

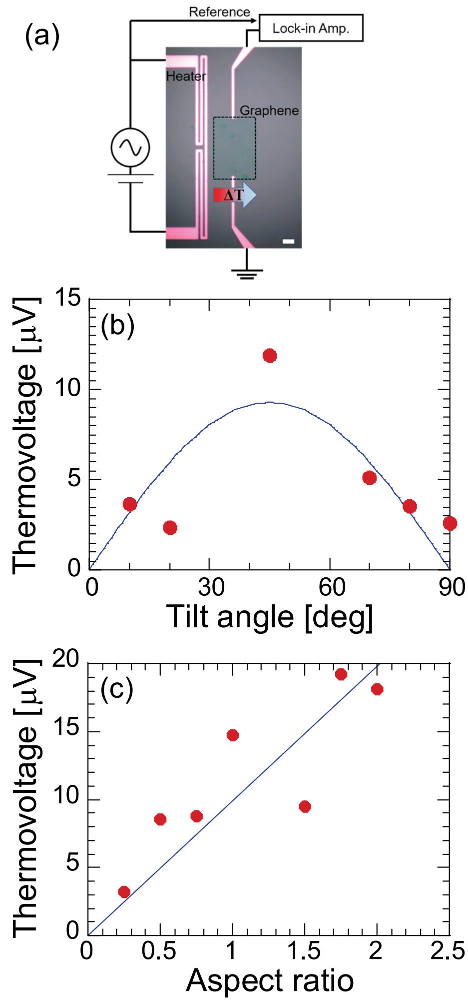

Fig. 4 (a) Transverse thermoelectric voltage measurement of graphene heterostructures. The measured thermovoltage as a function of (b) tilt angle and (c) aspect ratio of the device. Bar in (a) is 10 μm.