A light emitter based on polycrystalline graphene patterned directly on silicon substrates from a solid-state carbon source

Kenta Nakagawa^{1,2}, Hidenori Takahashi,² Yui Shimura² and Hideyuki Maki^{2,3}

 ¹ Kanagawa Institute of Industrial Science and Technology (KISTEC), 705-1 Shimoimaizumi, Ebina, Kanagawa 243-0435, Japan Phone: +81-46-236-1500 E-mail: mp-nakagawa@newkast.or.jp
² Department of Applied Physics and Physico-Informatics, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama, Kanagawa 223-8522, Japan. Phone: +81-45-566-1643 E-mail: maki@appi.keio.ac.jp
³ Center for Spintronics Research Network, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama, Kanagawa 223-8522, Japan Phone: +81-45-566-1643

Abstract

We developed a procedure for direct patterning of graphene with arbitrary position, size, and shape on silicon substrates from a solid-state carbon source without dry etching processing. Our light emitting graphene devices perform on a par with those based on high crystallinity graphene obtained via mechanical exfoliation or chemical vapor deposition.

1. Introduction

Nano-carbon materials such as carbon nanotubes and graphene are promising candidates for next-generation optoelectronic devices such as light emitters and detectors owing to their unique electronic and optical properties. Optical interconnects and silicon photonics are two key technologies for next-generation communication, and nano-carbon materials are regarded as one of the possible candidates for integrated optoelectronic devices because they can be easily integrated on silicon chips [1,2].

Graphene light emitters integrated on silicon chips are being developed as an alternative to conventional light sources based on compound semiconductors. Graphene light emitters based on the thermal radiation of graphene by transforming an electrical current into Joule heat are expected due to its (i) broad infrared emission by blackbody radiation, (ii) good compatibility to microfabrication technology, (iii) fabricating inexpensively on a silicon substrate, and (iv) direct modulation with a response speed of less than 100 ps (frequency 10 GHz) [2].

In general, graphene sheets are conventionally obtained by (i) mechanical exfoliation method or (ii) CVD. In the mechanical exfoliation method, graphene sheets are mechanically exfoliated from highly ordered pyrolytic graphite and transferred onto the substrates. In the CVD method, graphene sheets are synthesized by chemically reacting a hydrocarbon gas such as a CH_4 gas and a metal catalyst substrate such as a Cu single crystal at high temperature. Graphene sheets obtained by this method also require transferring the graphene from the conductive metal catalyst substrates to silicon substrates. Since both methods require a transferring process, graphene growth methods that allow for direct formation on silicon substrates have been sought after and several attempts have been reported.

In this study, we developed a procedure to grow polycrystalline graphene from a solid-state carbon source directly on silicon substrates. To reduce the complexity of the process, the number of steps, and the cost, we have also developed a direct patterning procedure that allows to produce graphene samples with arbitrary position, size, and shape. In addition, we have demonstrated that our polycrystalline graphene operates in light emitting devices as well as high crystallinity graphene obtained by mechanical exfoliation method or CVD [3].

2. Results and discussions

The procedure to grow graphene directly on silicon substrates from a solid-state carbon source is divided into three main steps: (1) vacuum vapor deposition, (2) rapid thermal annealing (RTA), and (3) Ni etching. In the vacuum vapor deposition step, Ni and amorphous carbon (a-C) were used as metal catalyst and solid carbon source, respectively. Ni (20 nm)/a-C (5 nm)/Ni (5 nm) stacked films were deposited on Si wafers by vacuum vapor deposition. In this study, RTA is utilized to synthesize graphene as an alternative to CVD. In the RTA tube, the substrates were heated up from room temperature to 1100 °C at a heating rate of +15 °C s⁻¹, and annealed at 1100 °C steadily for 2 min. A controlled descent back to room temperature was then performed at a cooling rate of -1 °C s⁻¹. After the RTA process, the substrates were immersed in an acid solution FeCl₃ (aq.) for about 24 hours to etch the metal catalyst Ni.

To reduce the process complexity, steps and cost for the integration on silicon chips, we also developed a direct patterning procedure that allows arbitrary positioning, dimensioning, and shaping of the graphene and that does not need dry etching of the sample afterwards. When the resist patterns were designed by conventional lithography before the RTA process; graphene position, size, and shape are controllable. Noteworthy, during the lithography step, only a very thin film of Ni has to be deposited on top of the silicon substrate to strengthen the adhesion of the Ni and the a-C stacked films to the substrate.

The optical microscope image of the directly patterned graphene is shown in Fig. 1(a). As shown in Fig. 1(a), graphene samples with various positions, sizes, and shapes were successfully grown. Since the position, size and shape of the graphene depend on the lithography, arbitrary shapes, such as spirals in Fig. 1(a), can be formed as well as simple square and rectangular shapes. The directly patterned graphene was characterized using Raman spectroscopy (Fig. 1(b)). The I_{2D}/I_{G} intensity ratio is about 0.953 indicating that the graphene is bi-layered. The existence of the D band indicates that the graphene is polycrystalline. Fig. 1(c) shows the D, G, and 2D band Raman mapping images obtained from a 20 μ m × 20 μ m square directly patterned graphene sample. The intensities of each of the Raman bands in the region are almost the same, indicating that the polycrystalline graphene is almost homogeneous.

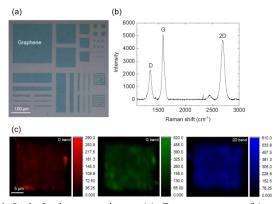


Fig. 1 Optical microscope image (a), Raman spectrum (b) and the D, G, and 2D band Raman mapping images (c) of the direct patterning graphene. Reproduced from Ref. [3] with permission from The Royal Society of Chemistry.

To test the viability of the directly patterned graphene for light emitting applications, Pd (145 nm)/Cr (5 nm) electrodes were deposited on the samples by vacuum vapor deposition as source and drain electrodes. The optical microscope image of the directly patterned graphene based light emitter is shown in Fig. 2(a). The size of the device graphene channels tested in this work was 5 μ m length and 5 μ m width.

The electrical and optical experiments were carried out at room temperature in a high-vacuum chamber. The infrared emission was observed with an InGaAs CCD camera sensitive to wavelengths between 0.9 and 1.6 μ m. Here, the emitted light was collected through the quartz optical window of the vacuum chamber and amicroscope lens. A bright emission from the directly patterned graphene between the two electrodes is observed under DC bias voltage ($V_{ds} = 9$ V) in vacuum. As shown in Fig. 2(b), the emission is highly localized in the graphene region. Fig. 2(c) shows the spectra of the light emitted by the graphene device. The broad spectra in the near IR region can be modelized with Planck's law, and the emission from the directly patterned graphene is considered blackbody radiation generated by Joule heating. The graphene temperatures, which are obtained by fitting Planck's law are estimated to be 748 K, 819 K and 920 K with $V_{ds} = 8$ V, 8.5 V and 9 V, respectively. These findings demonstrate that a light emitting device based on direct patterning graphene operates on a par with light emitting devices based on high crystallinity graphene obtained by mechanical exfoliation method or CVD [2].

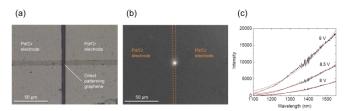


Fig. 2 (a) Optical microscope image of the direct patterning graphene light emitter. (b) IR camera image of the thermal emission at $V_{\rm ds} = 9$ V. (c) Emission spectra from the direct patterning graphene light emitter for $V_{\rm ds} = 8-9$ V with steps of 0.5 V. The red curves are the fitting results to Planck's law. Reproduced from Ref. [3] with permission from The Royal Society of Chemistry.

3. Conclusions

In conclusion, we developed a growth procedure for graphene directly patterned on silicon substrates from a solid-state carbon source. In addition, we also developed a direct patterning procedure that allows us to achieve graphene samples with arbitrary position, size, and shape without the need to undergo dry etching steps. Light emitting devices based on direct patterning graphene operate on a par with high crystallinity graphene based light emitting devices obtained by mechanical exfoliation method or CVD. Direct patterning graphene based light emitting devices integrated on silicon chips are expected to open new routes to the realization and commercialization of next-gen optical interconnects and silicon photonics. Moreover, applications are not limited to light emitting devices but can be extended to other optoelectronic devices such as photo-detectors and electronic device such as transistors, sensors, and circuits.

Acknowledgements

This work was partially supported by a project of Kanagawa Institute of Industrial Science and Technology (KISTEC), PRES-TO (Grant number JPMJPR152B) from JST, KAKENHI (Grant number 16H04355, 23686055, 18K19025, 20H02210 and 20K14783) and Core-to-Core program from JSPS, Spintronics Research Network of Japan, and NIMS Nanofabrication Platform in Nanotechnology Platform Project by MEXT.

References

- T. Mori, Y. Yamauchi, S. Honda and H. Maki, Nano Lett., 14 (2014) 3277.
- [2] Y. Miyoshi, Y. Fukazawa, Y. Amasaka, R. Reckmann, T. Yokoi, K. Ishida, K. Kawahara, H. Ago and H. Maki, Nat. Commun., 9 (2018) 1279.
- [3] K. Nakagawa, H. Takahashi, Y. Shimura and H. Maki, RSC Advances, 9 (2019) 37906.