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海外特別研究員最終報告書

独立行政法人日本学術振興会 理事長 殿

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(氏名は必ず自署すること)

海外特別研究員としての派遣期間を終了しましたので、下記のとおり報告いたします。
なお、下記及び別紙記載の内容については相違ありません。

記

1. 用務地（派遣先国名）用務地： アーヘン工科大学 （国名： ドイツ ）

2. 研究課題名（和文）※研究課題名は申請時のものと違わないように記載すること。
エッジ制御グラフェンナノリボンの革新的プラズマ合成手法の開発

3. 派遣期間：平成 30年 4月 1日 ～ 令和 2年 3月 10日

4. 受入機関名及び部局名
2nd Institute of Physics, Department of Physics, Faculty of Mathematics, Computer Science and Natural Sciences
RWTH Aachen University

5. 所期の目的の遂行状況及び成果…書式任意 **書式任意（A4判相当3ページ以上、英語で記入も可）**
（研究・調査実施状況及びその成果の発表・関係学会への参加状況等）
（注）「6. 研究発表」以降については様式10－別紙1～4に記入の上、併せて提出すること。

・派遣先における研究内容の設定と計画からの変更点

本派遣において，派遣先と研究の方向性について議論した結果，研究内容に変更があった．本研究テーマである，「エッジ制御グラフェンナノリボンの革新的プラズマ合成手法の開発」に向けて，当初の予定では派遣先においてグラフェンナノリボンの合成装置を立ち上げ，グラフェンナノリボンを合成し，電気伝導測定を行う予定であったが，派遣先で現在進行している類似した研究プロジェクトにおいて非常に興味深い研究結果が出始めていたため，その研究グループに加わり，研究を行うことになった．以降英語にて報告する．

1. Present project status

I belonged to the bilayer graphene quantum device team and was working on device fabrication. The concept of the device is based on valleytronics which utilizes a valley degree of freedom in the bilayer graphene. The desired function of the device is the valley splitter which can split the electrons which have deferent valley state (K, K*). We are planning to use a domain bond state which is shown in an interface between the two regions which are applied an opposite electric field in the bilayer graphene. To realize the fabrication of such device, I am trying to fabricate a well-designed graphite back-gate with high cleanness and accuracy.

2. Recent progress in fabrication

There were many problems in the fabrication process. Especially, overcoming the issue of the flipping process is most important to improve the device quality. Additionally, we are newly facing a bad effect on the transport of the device by flipping technique, especially by the annealing at high temperature (650℃)

when we remove PPC. We suppose that the annealing process causes movement of BLG encapsulated in hBN layers, this makes new alignment of BLG and hBN. It should be possible to create commensurate phase in BLG. We have already measured few devices, and these actually show the side peaks besides the Dirac-corn, this is the obvious sign of the presence of commensurate phase. With this, we cannot reach to pinch-off current because mini Dirac-corn emerged in the commensurate phase prevent to be dielectric. Therefore, the new approach to fabricate the kink-state device is highly required avoiding the annealing process to BLG and commensurate phase.

Furthermore, regarding the flipping technique, we developed the new process for fabricating the graphene nano-device. The base of this idea is placing the structured gate under the stack with an extremely clean interface. Combining to the top-gate and such structured bottom-gate, we can obtain the new freedom of the gating to the nano-graphene, making possible to freely tune Fermi level and even displacement field in arbitrary position. As the first challenge of the process for such device, we tried to employ finger gates under the BLG quantum point contact (QPC) formed with etching process.

Here I report the new fabrication process of the kink-state device with technical progress on the flipping process and the new process for the fabrication of graphene nano-device.

(A) New fabrication process of kink-state device

The main issue of the fabrication process we supposed is on annealing. Therefore, we tried to avoid the annealing process to BLG. For this, we arrived an idea to separately build a BLG sandwich and bottom part and combine these stacks in the end. Furthermore, we decided to employ the additional graphite global back gate to well tune the Fermi energy of the kink-state channel instead of the ordinal silicon global back gate. Thus, we need additional stacking process to place additional hBN and graphite. In this process, the annealing process is applied to only the bottom part, then we can completely remove the bad effect from annealing to BLG. The illustration of whole process is shown in Fig. 1. We assumed two routes of the process to arrive the full stack. One is the method using the alignment of the bottom split gate with PPC membrane when flipping it (Fig. 1(a)). To realize this way, precise handling of PPC membrane is necessary. The other way is picking up the separately prepared parts with BLG sandwich in order (Fig. 1(b)). In this method, we found that the gold structures, which are used when etching graphite, are still remained the substrate when we pick up the flipped bottom part. This phenomenon might happen if the gold structures are enough large because we have seen the tiny gold structures are successfully picked up with graphite. With this interesting phenomenon, we can remove the gold structures when picking up process. Therefore, we are finally able to obtain the full stack without gold structures.

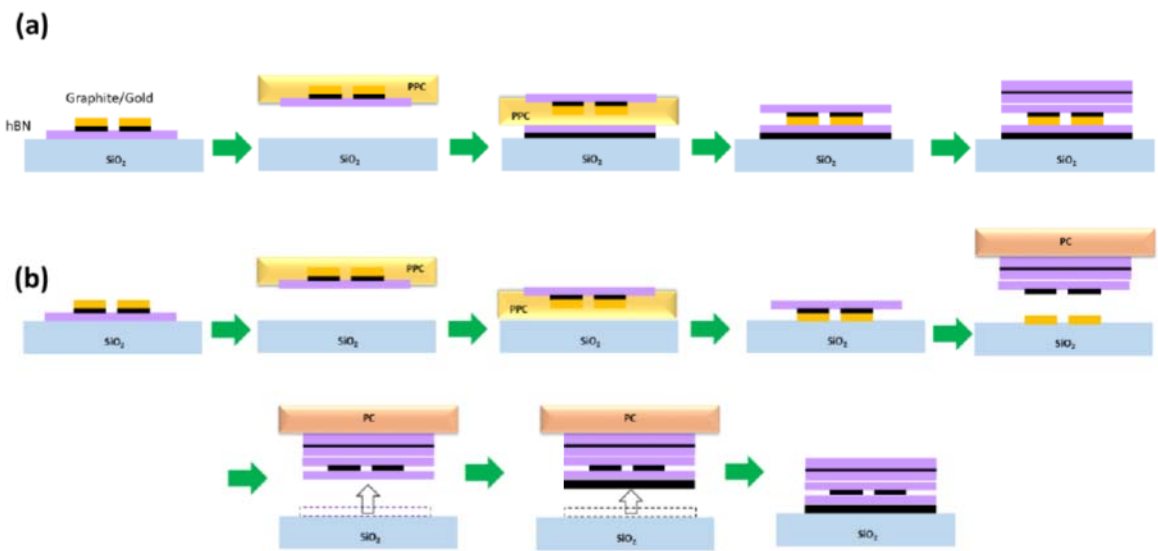

Fig.1: Two routes for the full stack. (a) and (b) shows the routes with and without alignment of PPC membrane.

To realize the process with the alignment of the PPC membrane regarding the process shown in Fig. 1(b), we improved the flipping method. In the previous method, we employed the PDMS stamp covered by tape as a support of the PPC membrane. In this way, however, the success yield of picking up is still quite low. Furthermore, the membrane easily ruptured when temperature goes higher. This also leads difficulty in handling the membrane. This might be because the membrane partially attaches to the substrate, resulting in rupturing the membrane. Therefore, we tried to directory put the PPC membrane on the substrate and peel off the membrane (Fig.2(a)). We annealed after putting the membrane (typically 50-100 ℃ for 1 min) and peel it off with the support of a tape. By this, we finally obtained the PPC membrane with picked up stack (Fig. 2 (b-c)). The membrane peeled off from the substrate has still flat surface, therefore we are easily able to handle it. Thanks to this, the membrane is possible to be suspended with the arm of the transfer system (Fig. 2(d)), this makes possible to align the stack on the graphite global back gate (Fig. 2(e,g)). After annealing to remove the PPC, we obtained two results, Fig. 2(f) shows good alignment of the split gate on the graphite global gate, meaning the stacks mostly did not move. On the other hand, we found the long-distance movement of the stack which can be seen in Fig. 2(h), this might happens due to the holdings of the membrane which is possible to cause the unexpected flow of PPC in annealing, resulting in the movement of the stack.

By these two process routes, we finally got two kind of stacks with and without gold structure (Fig. 3(a-b)). We can confirm that BLG was successfully aligned on the channel by raman mapping measurement (Fig. 3(c-d)). We will compare these two configurations in transport measurement, and clarify which configuration is suitable.

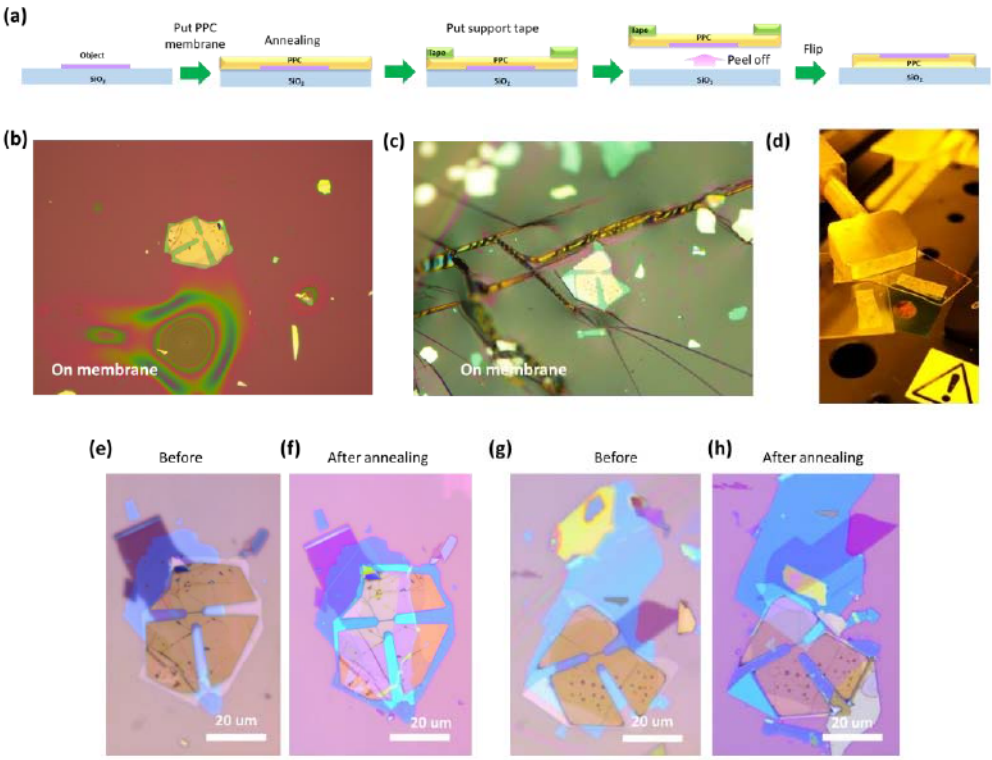

Fig.2: (a) The process of new flipping process with PPC. (b-c) The picked up bottom parts with PPC which has (b) flat surface and (c) holding. (d) A picture of tranfer system with suspended PPC membrane. (e, g) Pictures aligning the bottom part on the bottom global back gate with PPC. (g, h) Pictures after annealing of (e) and (g).

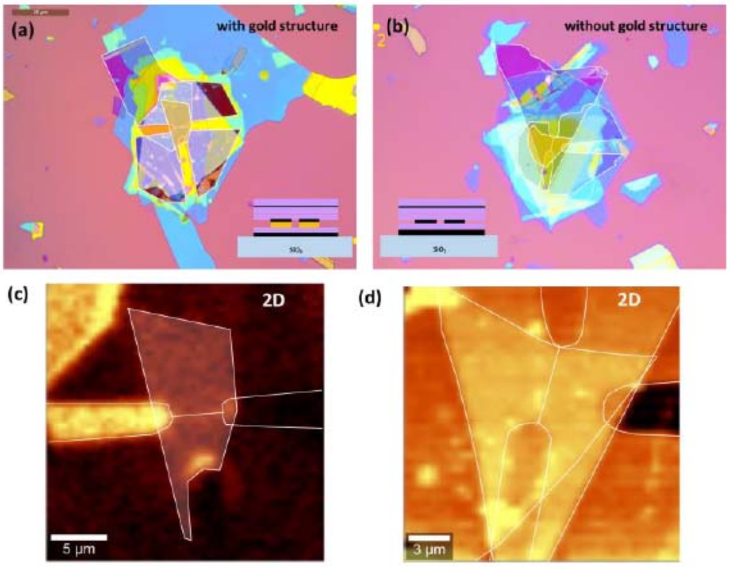

Fig.3: (a, b) Full stacks with nad without gold structures. (c, d) Raman mapping of 2D spectra of (a) and (b)

(B) New processing for nano-graphene device

Based on the flipping process we developed, an idea of new fabrication for a graphene nano-device came up. With the one of merit of the flipping technique that we can structure the bottom gate with clean interface, it is possible to place the structured bottom gate on a graphene nano-structure. The fabrication process is similar to that of kink state device, but deference is etching the graphene after flipping with gold hard mask. We expected this gold structure finally works like a top gate. Based on this technique, we envisioned the BLG quantum point contact (QPC) device with finger gates and top gate shown in Fig. 4(a-c). The split gates are apart from 150 nm each other and its width is 300 nm. BLG was etched with gold hard mask down to 200 nm. For this configuration, we expect the quantum dot (QD) operation because we can tune the displacement filed and thickness of tunnel barrier with finger gates. A device has already been ready for transport measurement (Fig. 4(d-e)). We measured the conductance in this device at low temperature (~1.5K). We found that this device showed the insulating state with BG and TG shown in Fig4 (a). This means this device is able to work as ordinal bilayer graphene device. This is actuary first report which shows the insulating state with etched bilayer graphene. Furthermore, we observed the periodic oscillation of conductance in pnp and npn regime. This imply the Fabry–Pérot oscillation which comes from the ballistic transport of carriers and that we realized really clean interface of hBN and BLG. In the such insulating regime, we further found the additional feature which is normally not observed in BLG shown in Fig. 4(b). This might be relating to the presence of etched edge of BLG. Such device configuration and measurement should improve the understanding of the impact of the edge to the graphene transport. This device is still in measurement and analysis in RWTH Aachen University.

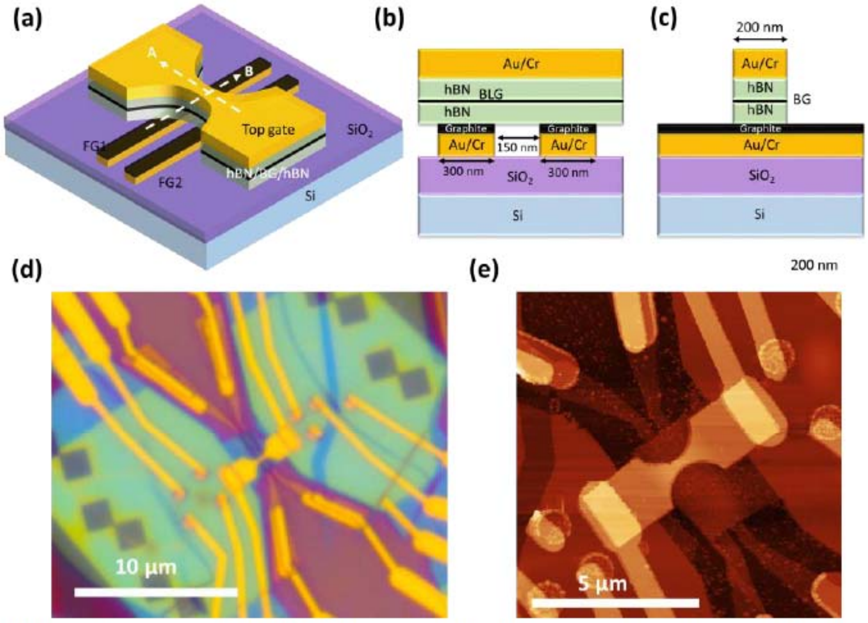

Fig.3: (a-c) Schematic images of BLG QPC device. (b)and (c) are cross section images along A and B direction shown in (a). (d) Optical microscope and (e) AFM image of the device.

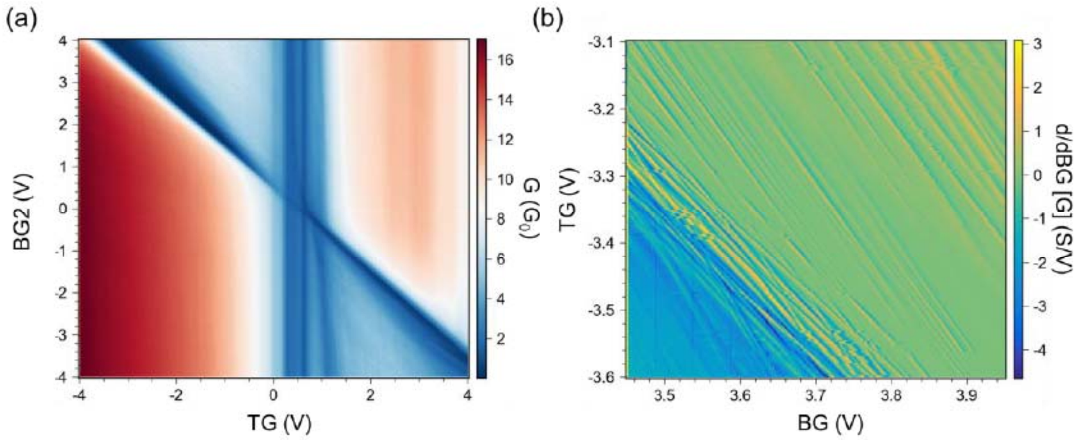

Fig.4: (a-b) The mapping of conductance with TG and BG2 (a) and TG and BG inside of the transport gap (b). The conductance in (b) is differentiative with BG.