Physisorption doping induced multiple dots behavior in graphene nanoconstrictions

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Abstract — **We report the single carrier transport properties in the p-doped/less-doped graphene nanoconstriction structures. In the doped graphene devices, the overlapped Coulomb diamond characteristics are observed around the charge neutrality point (CNP) at 5 K. Reducing doping in graphene by annealing, the periodic peaks appear in the certain gate voltage range around the CNP. Additionally, the non-overlapped Coulomb diamond characteristic is observed. These results suggest that unintentional charging island formation in graphene nanodevices can be avoided by decreasing the doping concentration.**

I. Introduction

Spin of electron in graphene quantum dots is a promising candidate for the implementation of the spin qubit in solid states [1]. Hence, the investigation of single carrier transport property in graphene is required. However, the single carrier tunneling at the inherent Dirac point (zero voltage) has not been observed due to unintentional doping in graphene [2]. Moreover, edge roughness and doping give rise to the uncontrollable formation of multiple charging islands in graphene, obstructing the single carrier transport operation [3]. To obtain the reliable single carrier property in graphene, it is imperative to examine its influence in detail. Here we study the doping effect on single carrier transport (Coulomb blockade) in graphene nanodevices.

II. Device fabrication and measurements

The devices were fabricated based on chemical-vapor-deposition-grown graphene on a heavily doped-Si substrate with the 300-nm-thick-SiO_2 layer, which was used as the back gate. By conventional electron beam lithography processes, Cr/Au metal contacts were fabricated. The graphene channel was patterned by plasma etching using a hydrogen silsesquioxane mask into the constriction structure (Fig. 1) to reduce the influence of edge roughness as much as possible. In order to investigate the doping effect, we compared the device characteristics before and after annealing, which shifts the CNP position (*i.e.*, doping concentration) [4]. Two terminal direct current measurements were performed in the vacuum of ~10^{-5} Pa at the temperature range of 5~300 K.

III. Results and discussion

In the gate characteristic before annealing shown in Fig. 2, the CNP was located in gate voltage $V_g > 40$ V at 300 K, meaning the heavily p-doped graphene owing to doping from the adsorption of oxygen and moisture, or remained water or contaminations at the interface between graphene and a substrate [4]. At 5 K, the oscillation characteristic was observed. The Coulomb diamond plot showed the overlapped conductance blockade region (Fig. 3). The charging energy is roughly estimated to be ~2.5 meV. These results can be attributed to the multiple charging islands formation in the constriction and source/drain regions.

Shifting the CNP to $V_g \sim 0$ V (after annealing), Coulomb oscillation was also observed around the CNP at 5 K (Fig. 4). In contrast to that before annealing, the periodic peak spacing of ~2.6 V was observed. In addition, the Coulomb diamond plot showed the non-overlapped blockade regions (Fig. 5), reflecting the single charging island formation in graphene. As the corresponding charging island size of ~538 nm^2 is smaller than the geometrical constriction region of ~2250 nm^2, therefore, the island is formed in the constriction region. The charging energy is increased to ~7.8 meV. These results indicate that the size and the number of charging island are varied with the doping concentration in graphene.

The single charging island behavior in the less-doped graphene constriction could be ascribed to quantum reflection at the end of the constriction, resulting in the quasi-bound state in the constriction region (Fig. 6). Importantly, it is implied that single carrier transistors with a single charging island could be realized in graphene by the combination of constriction structure patterning and CNP control. However, the asymmetric ambipolar curve at room temperature (Fig. 4(a)) and the variation in the size of Coulomb diamond (Fig. 5) were observed, that were caused by remaining potential inhomogeneity. This can be interpreted as that local doping distributed in graphene [5] persists even though the CNP is located at $V_g \sim 0$ V.

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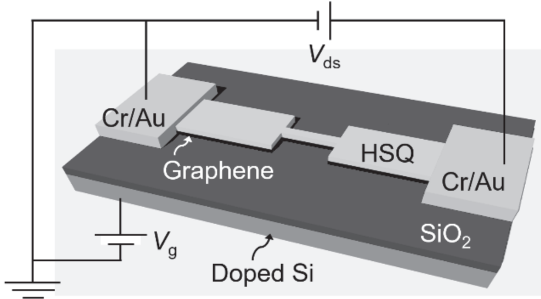

Fig. 1: Schematic illustration of the graphene constriction device and measurement configuration. V_{ds} and V_g are source-drain bias and back gate voltage, respectively.

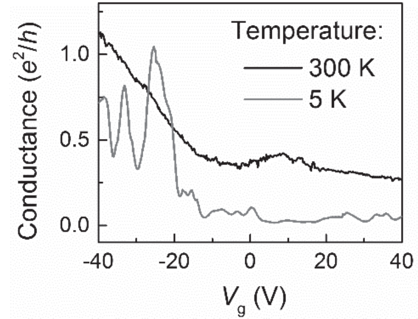

Fig. 2: Gate characteristic of the constriction device before annealing. Conductance is normalized with e^2/h (e: elementary charge, h: Planck constant).

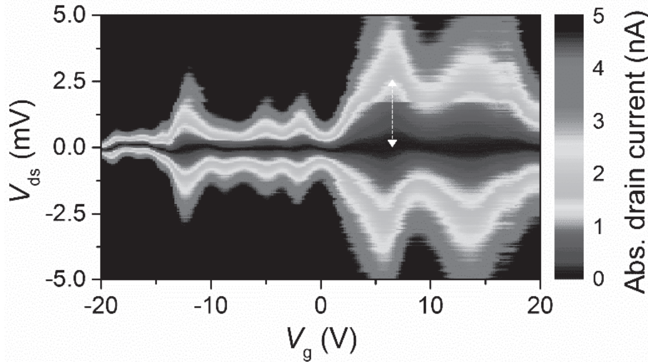

Fig. 3: Absolute drain current of the constriction device before annealing as a function of V_{ds} and V_g measured at 5 K. Charging energy is estimated on the dotted arrow.

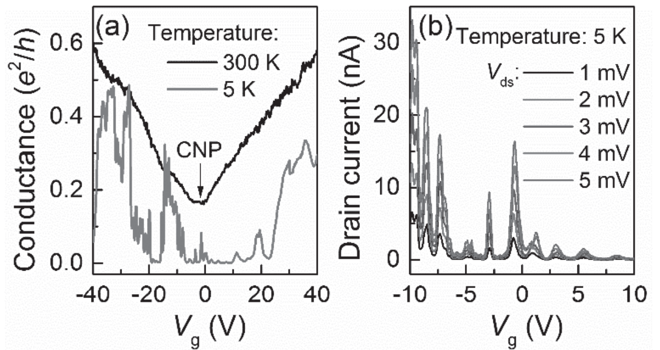

Fig. 4: Gate characteristic of the constriction device after annealing. (a) Conductance measured at 300 K and 5 K. (b) Drain current measured at 5 K applying various V_{ds}.

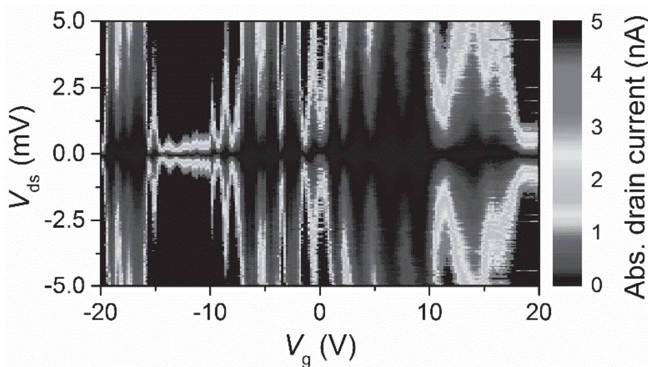

Fig. 5: Coulomb diamond plot of the constriction device after annealing measured at 5 K. Colour range is same as Fig. 3.

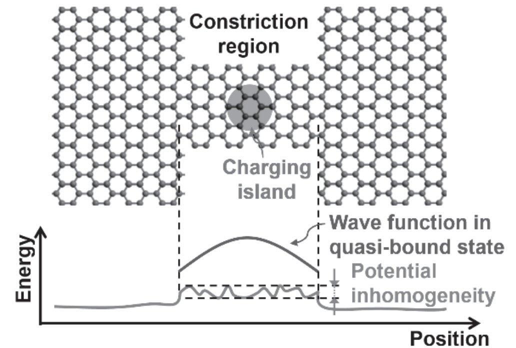

Fig. 6: Schematic energy diagram in the graphene constriction. Charging island is formed by quantum reflection at the end of the constriction region.