Growth mechanism of twisted graphene layers on graphene/silicon carbide template Osaka Univ.¹, Japan Atomic Energy Agency², NTT Basic Research Laboratories³ °Yao Yao¹, Ryota Negishi^{1*}, Daisuke Takajo¹, Yuki Fukaya², Makoto Takamura³, Yoshitaka Taniyasu³ and Yoshihiro Kobayashi¹ *E-mail: negishi@ap.eng.osaka-u.ac.jp

[Introduction] Twisted few layer graphene (FLG) has recently attracted great attention due to the appearance of the exotic electrical properties, such as superconductivity and Mott-like insulator [1]. In previous study [2], we reported that the FLG was synthesized by overlayer growth of graphene on a monolayer graphene template using a chemical vapor deposition (CVD) method. We found that the moiré pattern appears in the lattice structure of the grown graphene 2-dimensional (2D) island, indicating that the twisted angle between the grown graphene and template graphene is a value different from that of the order stacking. In this study, to understand the growth mechanism of twisted FLG, we examined the coverage dependence of vertical and lateral morphology in graphene islands grown on graphene template.

[Experiments] The 2D graphene islands with a monolayer step height were grown on silicon carbide with continuously oriented monolayer graphene via CVD with ethanol as a carbon feedstock. The growth was carried out in an infrared heating furnace at 1400°C [3]. The atomic force and scanning tunneling microscopes (AFM/STM) were used to evaluate the surface morphology and the twist angle. The interlayer distance between the grown graphene and template graphene layers was evaluated by the total-reflection high-energy positron diffraction (TRHEPD).

[Results and discussion] The AFM images (Fig. 1) show that coverage increases clearly when the growth time is prolonged. As coverage increases, the diameter of isolated graphene islands increase and the number of graphene islands decreases. This suggests that the plural small pieces of graphene are formed in a short period of time, and the large pieces of the graphene islands are formed by coalescence among the small islands via the lateral growth mode. The STM image with distinct moirés in Fig. 2 reveals the different rotation angle at several regions of the same large graphene island. It is verified that the large number of randomly oriented nuclei is formed grain boundaries via coalescence. Furthermore, the interlayer distance between the grown graphene and template graphene layers is evaluated to be 3.4 - 3.5 Å by the TRHEPD analysis. This value is obviously larger than 3.35 Å in the common AB stacking. We conclude that the grown 2D graphene islands have the random twisted angles due to the coalescence of islands, leading the formation of the weak interlayer coupling like a turbostratic stacking.



Fig. 1 AFM image and evaluation of grown 2D graphene islands

Number of graphene

island per 25 µm²

~1097

~156



Fig. 2 STM current images of grown 2D graphene island. (b)(c), enlarged images of the green square.

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