Supporting Information

Edge Control of Graphene Domains Grown on Hexagonal Boron Nitride

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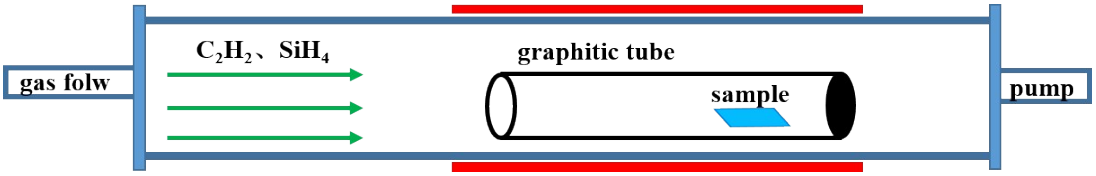

Figure S1. The schematic diagram of the CVD setup for graphene growth.

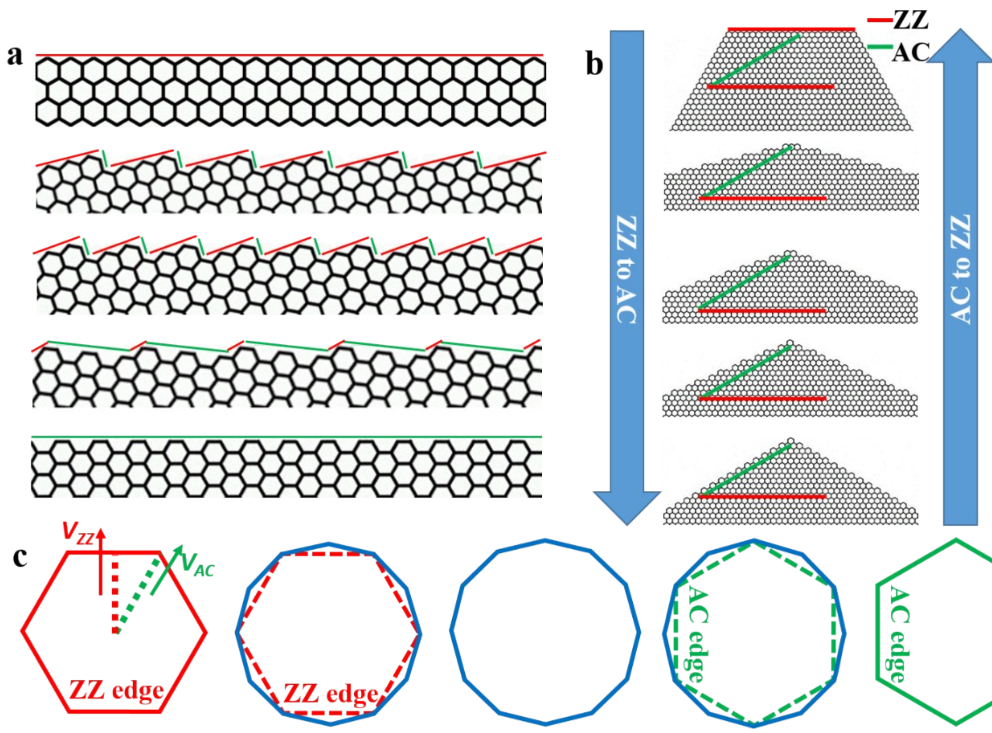

Figure S2. (a) illustration of edges with "pure" and mixed segments; (b) Edges of graphene domains having different ratios of ZZ and AC segments; (c) illustration of domain morphology varying from hexagon with "pure" ZZ-edges to dodecagon with high-ratio of ZZ segments, to regular dodecagon, to dodecagon with high-ratio of AC segments, and finally to hexagon with "pure" AC-edges. The red, green and blue lines represent "pure" ZZ segment, "pure" AC segment, and domain edges, respectively.

Fig. S2 illustrates the topographical evolution of single-crystal graphene domains. As shown in Fig. S2a, different edge is always of different ratios of AC and ZZ segments. Fig. S2b exhibits the evolution of neighboring edges in graphene domain, where segment-ratio alternates. This phenomenon could be well understood as different edge has different growth rate. As shown in Fig. S2c, assuming that AC and ZZ-edges have the same growth rates: $V_{AC}=V_{ZZ}$, where V_{AC} and V_{ZZ} are the growth rates of AC and ZZ-edges, respectively, the graphene domain is regular dodecagon. When $\sqrt{3}/2V_{ZZ}<V_{AC}<V_{ZZ}$, the graphene domain is dodecagon with high-ratio of AC segments. And when $V_{AC}\leq\sqrt{3}/2V_{ZZ}$, the graphene domain is hexagon with “pure” AC-edge. In contrast, when the growth rates of AC and ZZ are $\sqrt{3}/2V_{AC}<V_{ZZ}<V_{AC}$, $V_{ZZ}\leq\sqrt{3}/2V_{AC}$, the graphene domains are dodecagon with high-ratio of ZZ segments (Fig. 2F) and hexagon with “pure” ZZ-edge (Fig. 2E), respectively.

The growth rates of different oriented edges are determined by coefficient of catalysis of Si and etching of hydrogen. With the variation of ratio of ethyne and silane, different growth rates are obtained to get graphene domains with different edge structures. It also provides us a deep understanding about the mechanism for graphene growth with catalyst of silane.

NO.	T (°C)	Gas ratio (C_2H_2:SiH_4)	t (min)	P (Pa)	Topography	Growth rate (nm/min)
1	1300	8:0	15	2.58	hexagon of armchair	45
2		8:1	7	2.77	hexagon of armchair	65
3		8:4	5	3.30	hexagon of armchair	120
4		8:6	5	3.59	near circle	130
5		8:8	5	3.94	hexagon of zigzag	140
6		6:8	5	3.67	dodecagon	120
7		4:8	5	3.43	dodecagon	100
8		2:8	10	3.15	dodecagon	80
9		1:8	10	3.03	hexagon of armchair	55

Table S1. Summary of edge control experiments details under different growth conditions.

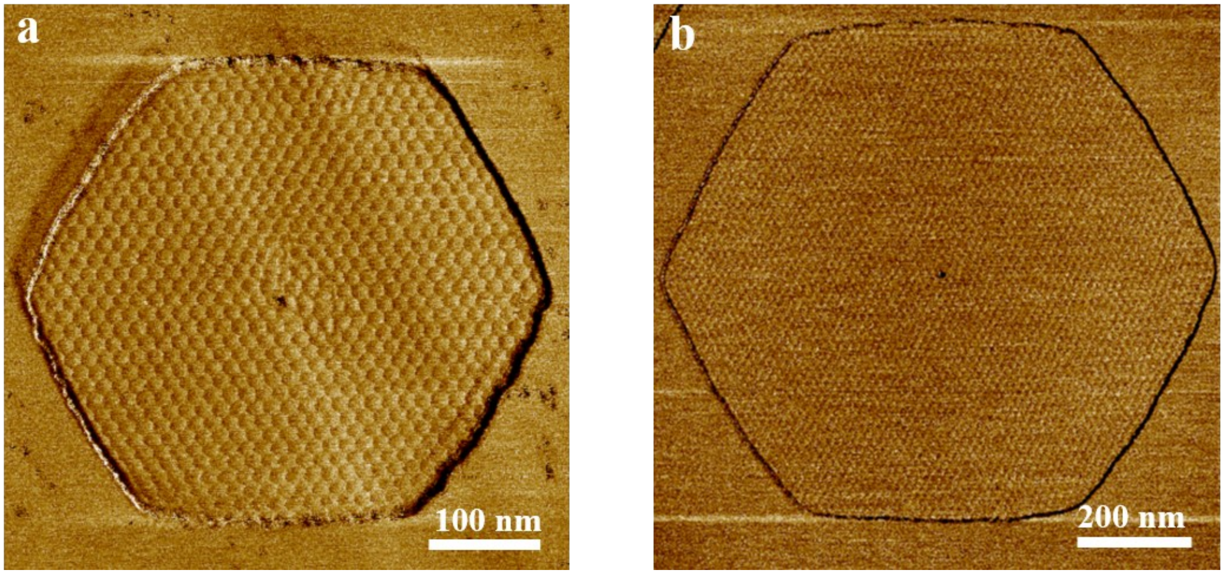

Figure S3. Graphene domains obtained in a gas flow of 8 sccm ethyne and 8 sccm silane with growth time of (a) 4 min and (b) 8 min, respectively. The edge orientation remains unchanged after the growth duration increases.

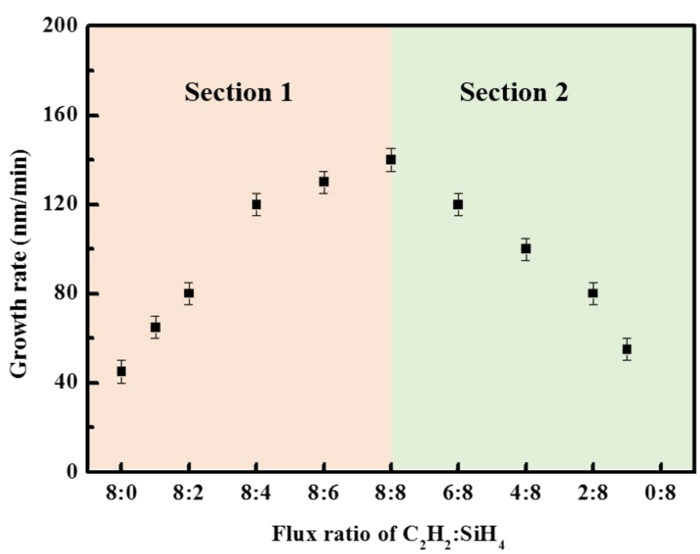

Figure S4. Section 1 represents the growth rates of supplying ethyne (C_2H_2) at 8 sccm and silane (SiH_4) from 1 sccm to 8 sccm in Fig. 2A-E, while section 2 represents the growth rates of keeping silane-flow at 8 sccm and ethyne-flow changing from 8 sccm to 1 sccm in Fig. 2E-I.