

Supplementary information

Switching isotropic and anisotropic graphene growth in a solid source CVD

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1. Optical microscope (OM) images of graphene domains for three different experiments.

OM analysis of the synthesized graphene on Cu foil was performed to measure the average graphene domain size as shown in the following figure S1. The OM analysis clearly shows the difference in graphene size for the three growth conditions. The average graphene domain size and growth speed of three different regions as shown in the OM images are calculated and included the following table S1.

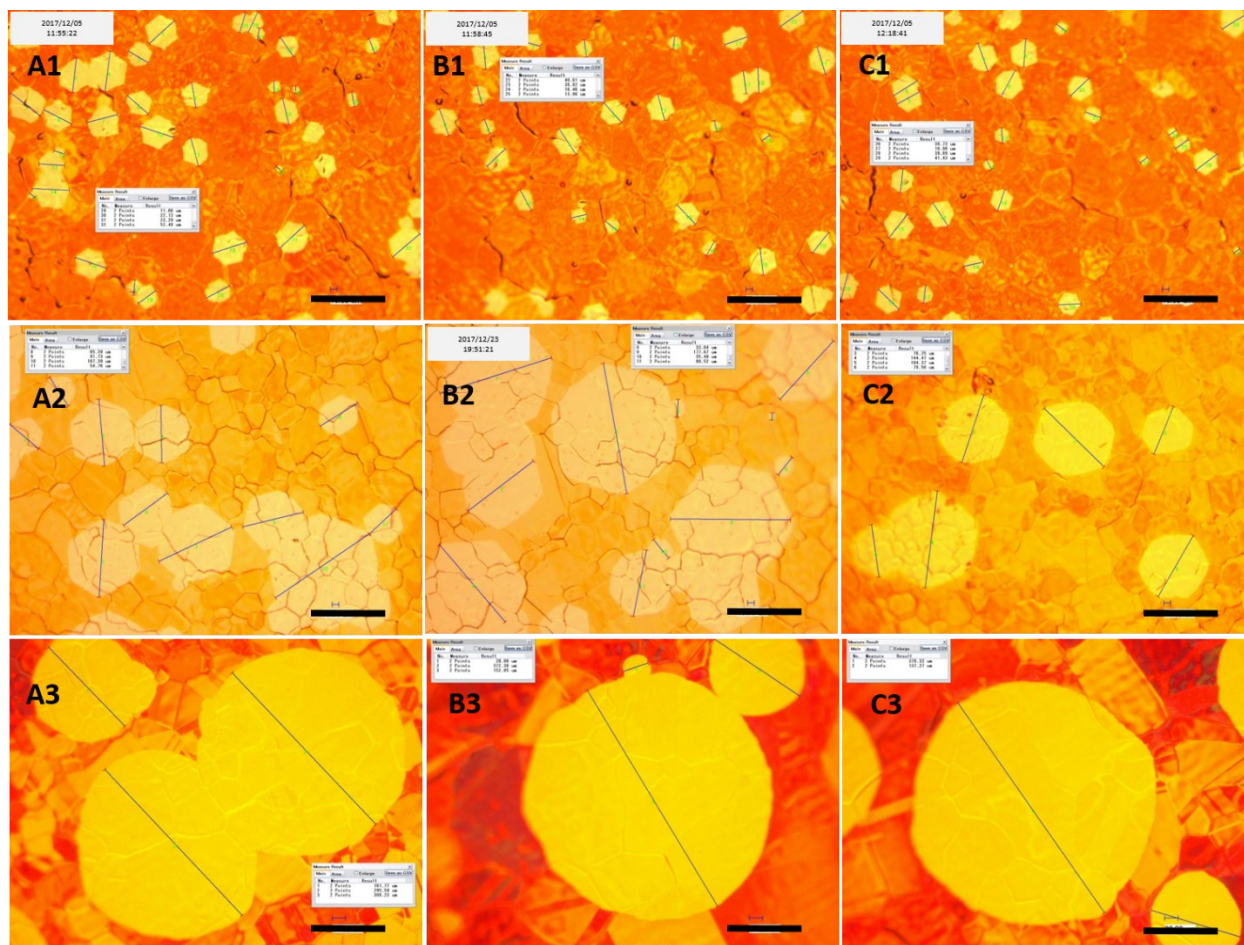


Figure S1. OM images of samples presenting the graphene domain sizes along their longest diagonal at three different regions of Expt 1 (A1-C1), Expt 2(A2-C2) and Expt 3. (A3-C3). Scale bars: 100 μm

Table S1. Calculation of average graphene domain size and growth speed of three different regions of three samples from the OM images of fig. S1.

Sample of Expt.	Region	No. of domains measured	Average Graphene domain size (μm)	Grand average (μm)	Average growth speed ($\mu\text{m}/\text{min}$)
1	A1	32	36.9059	33.7342	0.2765
	B1	25	32.8636		
	C1	29	30.9848		
2	A2	11	89.4536	98.4779	0.8072
	B2	11	103.7255		
	C2	6	105.4017		
3	A3	3	255.1967	230.5825	1.8900
	B3	3	187.8233		
	C3	2	257.8000		

2. Scanning electron microscope (SEM) images of graphene domains for three different experiments.

SEM analysis of the synthesized graphene domains on Cu foil was performed to measure the average domain density as shown in the following figure S2. The SEM analysis clearly shows the difference in graphene domain density for the three growth conditions. The average domain density of the three different regions as shown in the SEM images are calculated and included the following table S2.

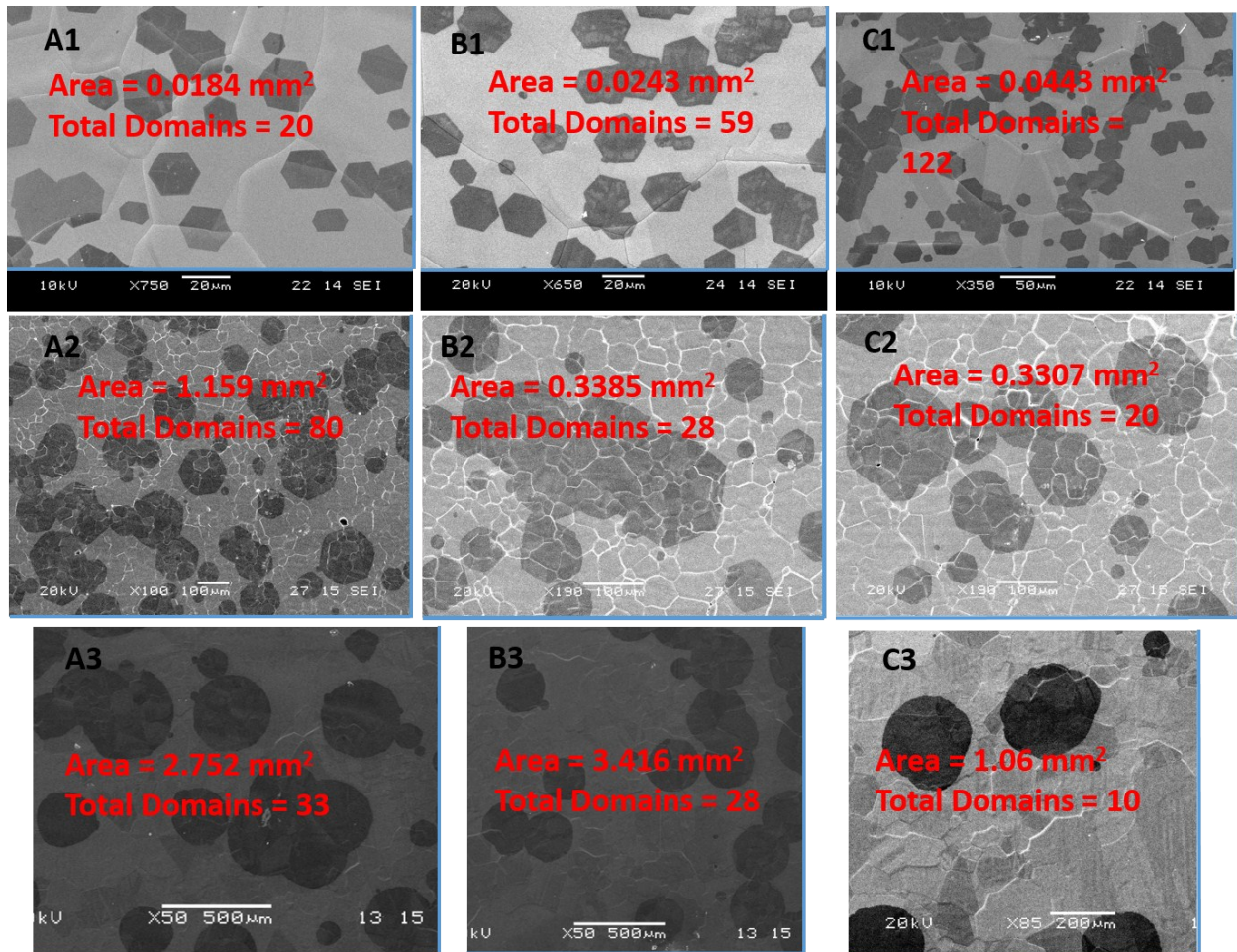


Figure S2. SEM images of three different regions of samples of (A1-C1) Expt 1, (A2-C2) Expt 2 and (A3-C3) Expt 3 with respective area and number of graphene domains counted.

Table S2. Calculation of graphene domains density at three different regions of three samples in figure S2.

Sample of Expt.	Region	Area(mm²)	Graphene Domains Count	Domains Density (mm⁻²)	Average (mm⁻²)
1	A1	0.0184	20	1086.9565	2089.6300
	B1	0.0243	59	2427.9835	
	C1	0.0443	122	2753.9503	
2	A2	1.1590	80	69.0250	70.7402
	B2	0.3385	28	82.7179	
	C2	0.3307	20	60.4778	
3	A3	2.7520	33	11.9913	9.8740
	B3	3.4160	28	8.1967	
	C3	1.0600	10	9.4340	

3. Cu domain size by optical microscope (OM) analysis

The OM analysis of the annealed Cu foil without synthesizing the graphene shows the distribution of Cu grain. A variation of Cu grain with the three growth conditions can be observed from the annealed sample as shown the following figure S3. In table S3 the calculated of average Cu-grain sizes at three different regions of three samples are summarized.

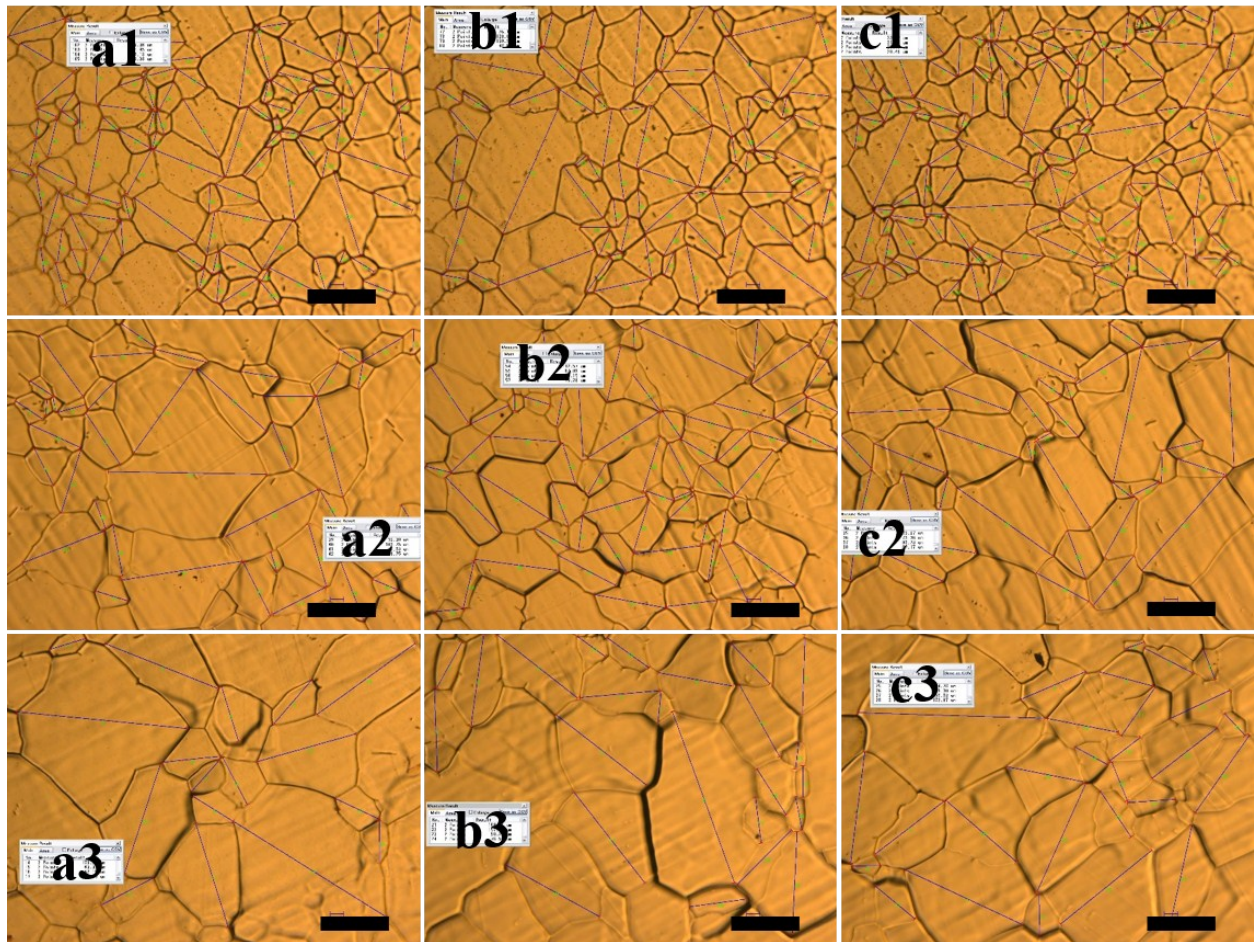


Figure S3. OM images of annealed samples to determine Cu grains size at three different regions of (a1-c1) Expt 1, (a2-c2) Expt 2 and (a3-c3) Expt 3. (Scale bars: 100 μm).

Table S3. Calculation of Average Cu-grains sizes at three different regions of three samples in fig. S3.

Sample of Expt.	Region	No. of Grains	Average Cu-grain size (μm)	Grand Average (μm)
1	a1	123	50.3309	54.8547
	b1	80	63.0086	
	c1	105	53.9414	
2	a2	42	79.3105	79.8343
	b2	57	74.0811	
	c2	38	89.0429	
3	a3	17	122.6188	110.5355
	b3	24	108.2763	
	c3	28	105.1357	

4. Electron back scattered diffraction (EBSD) analysis

The EBSD analysis was performed to observe the crystallographic nature of the annealed Cu foil for the three different growth conditions. The EBSD result can be correlated with the previous results of variation in Cu grain with change in volumetric amount of gas flow in the three growth conditions.

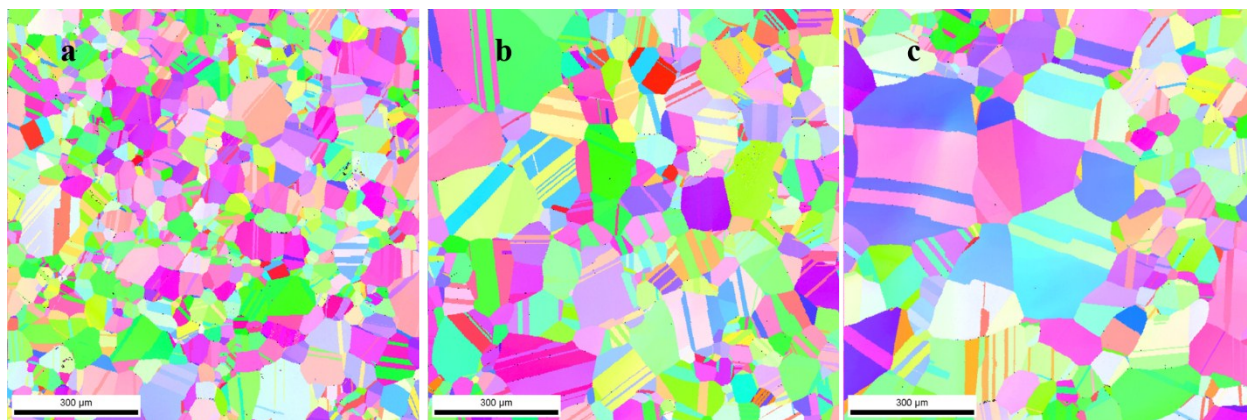


Figure S4. EBSD images of annealed samples revealing the comparative Cu grains size of (a) Expt 1, (b) Expt 2 and (c) Expt 3.

5. Raman point spectra analysis for the three different graphene domains

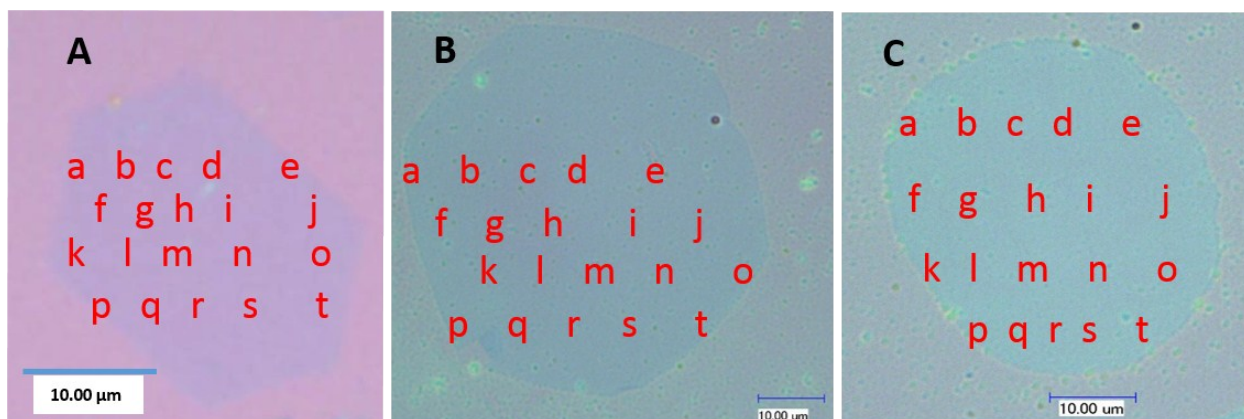


Figure S5. OM images of transferred graphene domains on SiO_2/Si of (A) Expt 1, (B) Expt 2 and (C) Expt 3 with 20 points at which Raman spectra are taken for D/G and 2D/G ratios.

Table S4. Calculation of D/G and 2D/G peak ratio of Raman spectra for the hexagonal domain as of figure S5 (A).

Position of spectra	D-Peak height (au)	G-Peak height (au)	2D-Peak height (au)	D/G peak Ratio	2D/G peak Ratio
a	26.33	256.54	557.54	0.1028	2.1777
b	9.19	239.76	539.65	0.0384	2.2507
c	4.21	262.99	601.94	0.0160	2.2888
d	9.03	230.56	547.04	0.0392	2.3727
e	8.09	238.62	553.35	0.0339	2.3189
f	12.20	249.15	551.87	0.0490	2.2150
g	6.47	254.94	550.31	0.0254	2.1585
h	4.84	232.82	529.69	0.0208	2.2751
i	5.06	240.47	550.77	0.0210	2.2904
j	5.33	249.44	561.06	0.0214	2.2493
k	5.57	245.55	523.82	0.0227	2.1333
l	5.29	253.54	530.82	0.0217	2.0936
m	3.49	218.07	466.82	0.0160	2.1407
n	4.17	231.05	523.15	0.0181	2.2642
o	5.12	247.51	514.39	0.0207	2.0783
p	6.35	243.71	545.16	0.0261	2.2369
q	20.06	243.57	552.93	0.0824	2.2701
r	5.60	243.88	526.84	0.0230	2.1602
s	4.11	244.37	542.06	0.0168	2.2182
t	5.03	252.75	507.85	0.0199	2.0093
Average				0.0317	2.2101

Table S5. Calculation of D/G and 2D/G peak ratio of Raman spectra for the imperfect hexagonal domain as of fig S5 (B).

Position of spectra	D-Peak height (au)	G-Peak height (au)	2D-Peak height (au)	D/G peak Ratio	2D/G peak Ratio
a	8.19	141.45	459.02	0.0579	3.2451
b	8.97	153.55	461.19	0.0584	3.0035
c	7.00	153.55	463.42	0.0457	3.0345
d	19.27	149.62	436.30	0.1288	2.9161
e	5.47	163.94	421.71	0.0333	2.5724
f	10.71	202.89	404.43	0.0528	1.9933
g	8.72	168.73	412.28	0.0517	2.4434
h	8.56	169.29	394.39	0.0506	2.3297
i	14.96	170.31	408.85	0.0879	2.4007
j	6.32	166.41	391.19	0.0380	2.3508
k	6.71	167.37	390.72	0.0401	2.3345
l	7.25	169.94	397.81	0.0427	2.3409
m	11.44	148.24	386.90	0.0772	2.6100
n	6.75	167.41	395.53	0.0404	2.3627
o	8.82	173.90	405.49	0.0507	2.3317
p	5.70	172.46	386.84	0.0331	2.2431
q	11.66	151.94	488.04	0.0767	2.9488
r	7.42	157.77	405.91	0.0470	2.5728
s	7.34	157.05	416.56	0.0468	2.6525
t	6.61	171.10	393.08	0.0386	2.2974
Average				0.0549	2.5492

Table S6. Calculation of D/G and 2D/G peak ratio of Raman spectra for the circular graphene domain as of figure S5 (C).

Position of spectra	D-Peak height (au)	G-Peak height (au)	2D-Peak height (au)	D/G peak Ratio	2D/G peak Ratio
a	123.00	2037.03	6620.49	0.0600	3.0200
b	84.02	2090.32	6596.06	0.0400	3.1500
c	85.06	2068.95	6911.76	0.0400	3.3400
d	237.22	2071.31	6719.31	0.1100	3.0240
e	86.11	2107.95	6664.94	0.0400	3.1600
f	73.22	1967.85	6661.55	0.0370	3.3800
g	70.35	2137.13	6097.07	0.0330	2.8500
h	130.19	2106.54	6321.04	0.0610	3.0007
i	76.97	2177.30	6904.94	0.0350	3.1710
j	170.23	2152.20	6718.25	0.0790	3.1200
k	207.79	2107.92	6565.71	0.0980	3.1140
l	89.38	2058.46	6368.83	0.0430	3.0930
m	51.75	2170.98	6048.15	0.0240	2.7850
n	122.17	2109.86	6433.83	0.0580	3.0490
o	138.19	2165.16	6602.20	0.0640	3.0490
p	141.16	2056.31	6455.67	0.0680	3.3190
q	47.84	2267.83	6468.70	0.0210	2.8520
r	140.46	2126.67	6519.18	0.0660	3.0650
s	97.03	2123.48	6751.75	0.0457	3.1760
t	98.59	2181.38	6915.42	0.0450	3.1700
Average				0.0534	3.0944