Growth of embedded and protrusive graphene rings on 6H-SiC (0001) by thermal decomposition in argon gas atmosphere

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Optical antennas are devices that efficiently convert the energy of free propagating radiation into localized energy and vice versa. The development of optical antennas can improve the efficiency of sensing, photodetection, light emission, spectroscopy and so on. Graphene rings are promising material for developing optical antennas because its plasmons have more electromagnetic confinement than metallic plasmons.

We grew epitaxial graphene by employing N-type Si-terminated 6H-SiC (0001) substrates. The sample was first cleaned by ultrasonic precleaning with acetone then mounted on the sample holder and put in a main chamber with the base pressure of ~10^{-10} mbar. In order to remove oxide on the sample's surface, we deposited silicon around 2 monolayers on it. These silicon atoms are expected to react with oxide molecules and leave the surface in the form of SiO_x when the sample was annealed at high temperature. Then the procedure for annealing SiC started by resistive heating in this UHV chamber. In case of annealing SiC substrate under Ar gas the sample had been transferred, without exposure to air, to another annealing chamber before annealed them by resistive heating under an Ar gas pressure of 0.01-0.5 atm. The annealing temperature was in range of ~900 °C to graphitization temperature (1350 °C - 1700 °C) with steps of ~100 °C (10~15 min per each step). The annealing temperature was measured by an optical pyrometer.

After graphene was grown on the SiC substrates, the graphene morphology and shape on all samples are measured by AFM. We find that graphene rings can be grown under some conditions. Figure 1 shows a final annealing temperature and ambient Ar pressure graph displaying the condition which graphene rings can be grown on the SiC substrates. We found that graphene ring can be grown under the condition of Ar pressure of 0.05-0.1 atm (as indicated by triangle symbols). At this Ar pressure and the final annealing temperature of 1550 °C, embedded graphene rings can be grown on the samples (Figure 2 (a)-(c)). Under the condition of Ar pressure of 0.05 atm, we also try to anneal the substrate with higher temperature (1650 °C). Surprisingly, the protrusive graphene rings are found on the sample as shown in Figure 2 (d)-(f).

Figure 2 shows AFM topography and phase images of the samples which annealed under Ar pressure of 0.05 atm with final annealing temperature of 1550 °C (Figure 2 (a)-(c)) and 1650 °C (0.05 atm 1650 °C sample) (Figure 2 (d)-(f)) and Ar pressure of 0.3 atm with the final temperature of 1675 °C (Figure 2 (g)-(i)). In the case of 0.05 atm 1550 °C sample (Figure 2 (a)-(c)), AFM phase image (Figure 2 (b)) shows there are many graphene rings on this sample (the bright regions indicate graphene regions). The AFM topographic image (Figure 2 (a)) shows the position of graphene rings is lower than that of SiC on the same terrace. Figure 2 (c) displays the magnification of the morphology of graphene ring in the rectangle in Figure 2 (a) reveals the graphene ring is embedded in SiC surface with the depth of 0.82 nm. For annealing with higher temperature (0.05 atm 1650 °C sample), there are also many graphene rings are observed on the sample surface as shown in Figure 2 (e). The C diffusion length and terrace width of this sample in average are about 1.2 μm and 2-2.5 μm, respectively. The AFM topographic image (Figure 2 (d)) reveals surprised results that the growth of graphene rings under this condition gives us protrusive graphene rings. It is opposite to the case of 0.05 atm 1550 °C sample (Figure 2 (a)-(c)) which showing the embedded graphene rings. Figure 2 (f) shows the magnification of a protrusive graphene ring image where the bright hollow hexagon is graphene region. This graphene ring is about 5.8 nm and 8.3 nm higher than the SiC terrace and pit inside, respectively. In the case of 0.3 atm 1675 °C sample, the average of terrace width on this sample is about 1.25 μm. There are only graphene stripes (no graphene island) on the terraces suggests that the C diffusion length is larger than the terrace width (~1.25 μm). Figure 2 (g) and (h) are AFM topographic and phase image, respectively measured at position which contains wide terrace of about 3-6 μm. Protrusive graphene islands are found on the wide terrace. Figure 2 (i) shows the magnification of morphology of protrusive graphene island in the rectangle in Figure 2 (g) reveals that protrusive graphene islands with the height of 2.2 nm have no pit inside. It is different from graphene rings on the 0.05 atm 1650 °C sample (Figure 2 (d)-(f)) which has a deep pit inside.

We found the growth of embedded and protrusive graphene rings on Si-terminated 6H-SiC (0001) by annealing SiC substrates under Ar gas pressure of 0.05-0.1 atm. The type of graphene rings (embedded or protrusive graphene rings) depends on the annealing temperature. The highest density of embedded and protrusive graphene rings occurs after annealed the SiC substrates under Ar pressure of 0.05 atm with annealing temperature of 1550 °C and 1650 °C, respectively.

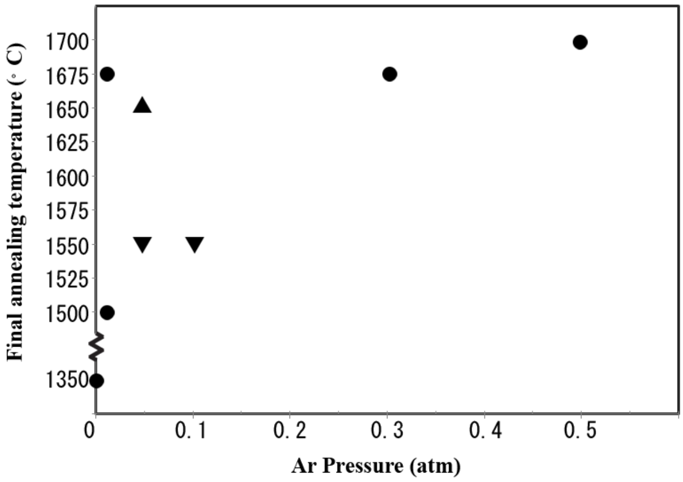

Figure 1 Final annealing temperature vs Ar pressure condition for the growth of epitaxial graphene on 6H-SiC (0001). triangles and Circles indicate the presence and absence of graphene rings, respectively. A point-up and point-down triangles indicate protrusive and embedded graphene rings on the samples, respectively.

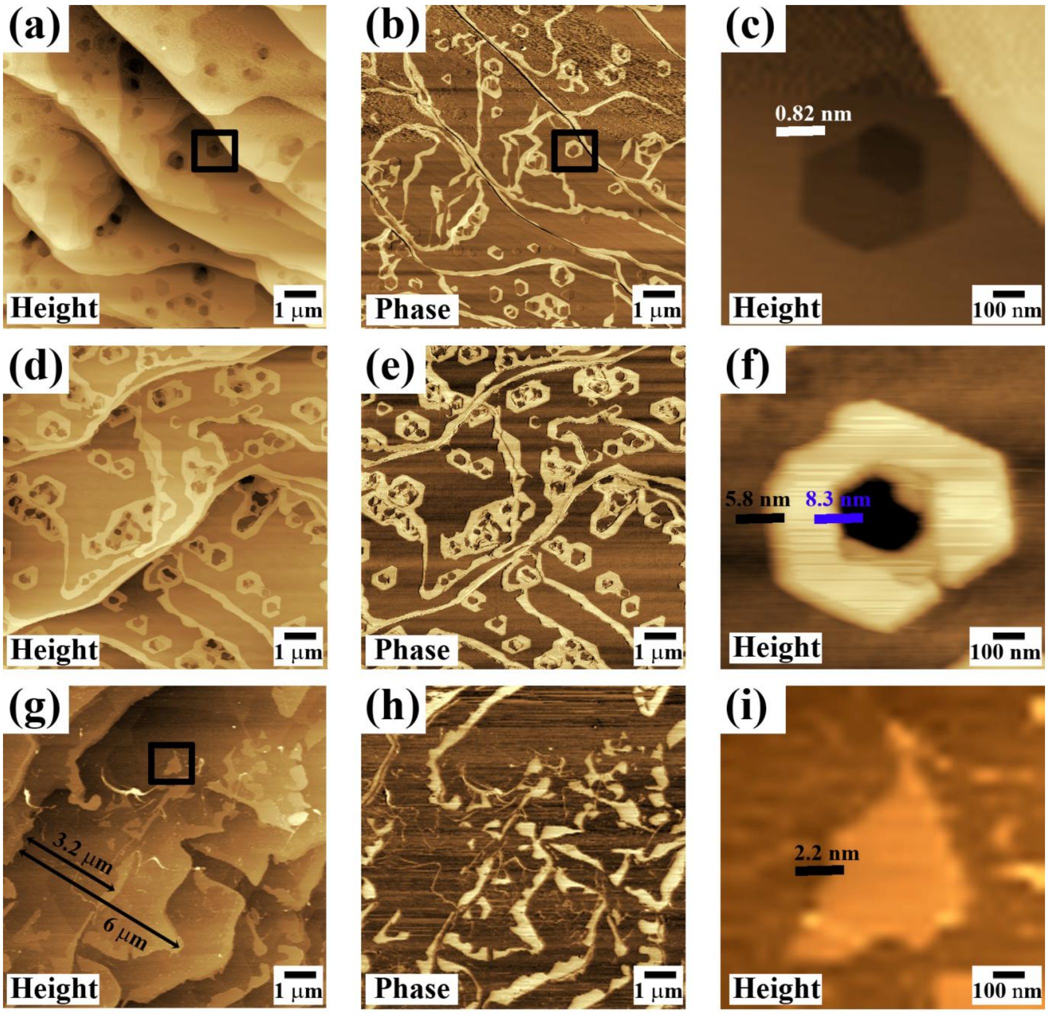

Figure 2 AFM images of the sample which annealed under Ar pressure and final annealing temperature of 0.05 atm, 1550 °C ((a)-(c)) and 0.05 atm 1650 °C ((d)-(f)) and 0.3 atm, 1675 °C ((g)-(i)). AFM mode type is labeled inside the image (height or phase). (c) and (i) Magnification of the square in (d) and (g), respectively.