

Determination of epitaxial graphene thickness by X-ray diffraction

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Introduction

Graphene is an allotrope of carbon created by arrangement of carbon atoms in a two-dimension honeycomb lattice. The unit cell of graphene, in which two carbon atoms are contained, is formed by two lattice vectors, $|a_G| = |b_G| = 2.4589 \text{ \AA}$. Graphene has two important bonding which are π bond perpendicular to the planar sheet and sp^2 bond (σ bond) in-plane sheet.

Graphene can be grown by many ways such as mechanical exfoliation of graphite, chemical vapor deposition (CVD) of carbon-bearing gases on the surface of copper films, and cutting open nanotubes. Annealing SiC is also one of the efficient approaches for a large scale production of graphene. There are several techniques, such as Auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS) and Ellipsometry, to determine the layer number of graphene grown on SiC. The accuracy of their techniques depends on their models. In this report, the estimation of layer number of epitaxial graphene is studied by using the direct information from X-ray diffraction (XRD). The result can give us the accurate number of epitaxial graphene layers.

Experiment

N-type Si-terminated 6H-SiC(0001) substrates of $12 \times 3 \times 0.25 \text{ mm}$ were employed for the graphene growth. The width and longitudinal orientations of the substrate are $[-1, 0, 1, 0]$ and $[-1, 2, -1, 0]$, respectively. The sample was first cleaned by ultrasonic precleaning with acetone. After the acetone was evaporated from the sample's surface, the sample was immediately mounted on the sample holder and put in the main chamber with the base pressure of $\sim 10^{-10} \text{ mbar}$. In order to remove oxide on the sample's surface, we deposited silicon around 2 monolayers on it. The deposited Si molecules are expected to crash and eliminate the oxide molecules from the sample surface. Then the sample was transferred, without exposure to air, to an argon chamber where it was annealed by resistive heating in an argon pressure of 0.3 atm. The annealing temperature was in a range from ~ 900 to $\sim 1675^\circ\text{C}$ with steps of $\sim 100^\circ\text{C}$ (~ 10 - 15 min per each step). The annealing temperature was measured by an optical pyrometer.

The XRD measurement was carried out at beam line 3A and 4C. Diffraction data were collected at X-ray energy of 10.2 keV.

Result and Discussion

Figure 1 (blue dots) shows X-ray diffraction (XRD) profiles. We can observe the graphene peak at ~ 20.9 degree and SiC peak at ~ 28 degree for $\lambda = 1.21553 \text{ \AA}$. The d-spacings of the two peaks due to graphene and SiC are $\sim 3.35 \text{ \AA}$ and $\sim 2.51 \text{ \AA}$, respectively.

The XRD profiles can moreover give us the graphene thickness information accurately. In order to estimate the graphene thickness, a distribution of the graphene thickness on SiC is required to be modeled. We assume that our sample contains the mixture of graphene monolayer, bilayer and trilayer. In this model, we assume that graphene on SiC is separated to monolayer, bilayer and trilayer regions. The proportion of each region is 15%, 45% and 40%, respectively. At each part, the presence of atoms above the topmost layer decrease exponentially. The XRD intensity for this model is

$$|F|^2 \propto \left| \sum_0^{l=N} P_l \sum_0^{j=l} \beta_j^j e^{ikaj} \right|^2$$

Where $ka = (4\pi d \sin\theta)/\lambda$; N is the maximum of graphene layer number on SiC substrate, β_j is the occupancies of j^{th} graphene layer in P_l region (the range of β_j is $0 \leq \beta_j \leq 1$), P_l is the relative population. A red curve in Figure 1 shows the best fit curve obtained from the above equation by using the parameter $\beta_1 = 0.90$, $\beta_2 = 0.55$, $\beta_3 = 0.75$, $P_1 = 0.15$, $P_2 = 0.70$ and $P_3 = 0.15$. The relative area of monolayer, bilayer and trilayer can be calculated to be 34%, 23% and 6.0%. The average is $(0.34 + 2 \times 0.23 + 3 \times 0.06) / 0.63 = 1.6$ graphene layers.

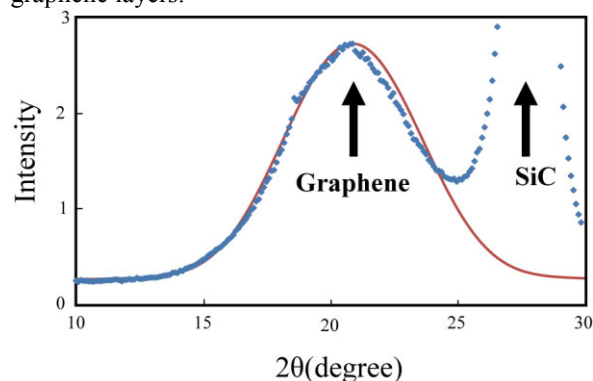


Figure 1. Calculated curve (red curve) fits on the XRD profile (blue dots).

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