Electronic Supplementary Material

Growth of Large-Area Single- and Bi-Layer Graphene by Controlled Carbon Precipitation on Polycrystalline Ni Surfaces

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1. Calibration of {*G*} with the number of layers using HOPG-derived graphene samples

We use the color contrast generated by 1-, 2- and 3-LG derived from exfoliated HOPG and deposited on 300 nm SiO_2/Si as an automated calibration for the assignment of the number of layers of the CVD graphene films (also on 300 nm SiO_2/Si) as described below. Figure S-1(a) shows microcleaved graphene pieces on 300 nm SiO_2/Si. The number of layers can be determined by inspecting the *G*´ peak (at ~2700 cm^{-1}) in the Raman spectra of the graphene pieces (Fig. S-1(b)). AFM can also assist in the layer number assignment (Fig. S-1(c)). The RGB (red green blue) color model is a model used for displaying and representing optical images (Foley, J. D. *Computer Graphics: Principles and Practice;* Addison-Wesley: Reading, Mass. 1996) and is shown here to be useful for automated assignment of the layer number. In this model, each pixel of an image mixes red, green, and blue light to reproduce the color of a pixel. The color obtained for each pixel depends on the intensities of the red, green, and blue components that are mixed. Figure S-1(d) shows the values corresponding to the green component {*G*} extracted from the optical image along the line in Fig. S-1(a). The red, green, and blue values of each pixel of our images are expressed on a scale of 0–255 (8-bit per channel). A stepwise change of {*G*} is observed (Fig. S-1(d)) with respect to the value of {*G*} corresponding to the pixels of the bare SiO_2/Si substrate. Each step in Fig. S-1(d) corresponds to the addition of one graphene layer. In Fig. S-1(d), the

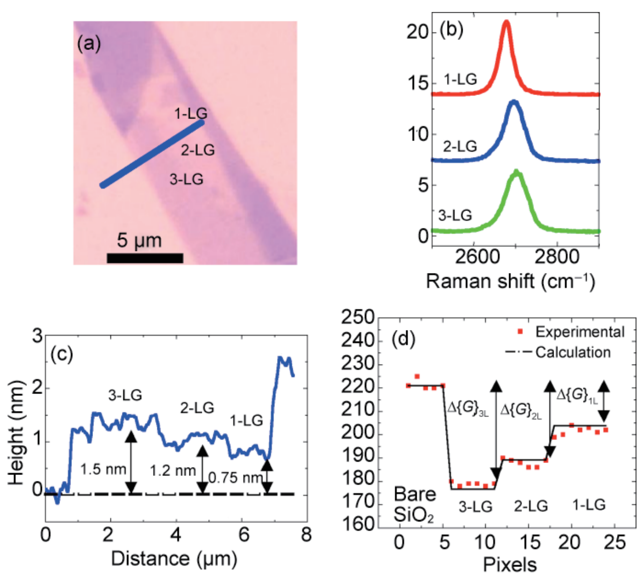


Figure S-1 (a) An optical image of microcleaved HOPG graphene (1–3 layers) on 300 nm SiO_2/Si; (b) Raman spectra of the regions identified as one, two or three graphene layers in (a); (c) measured AFM cross sectional height vs distance corresponding to the blue line in (a); (d) {*G*} values extracted from the RGB values along the blue line shown in (a). The black line shows the calculated {*G*} values for one, two, and three graphene layers on SiO_2 and the red points are experimental readings black line shows the calculated decrease in {G} with respect to the bare SiO_2 {G} value. This is obtained by calculating the reflectivity of bare SiO_2/Si and graphene on SiO_2/Si at a wavelength of 532 nm (see discussion below). We define the difference between {G} of graphene layers on SiO_2/Si and {G} of a bare SiO_2/Si substrate as Δ{G} (see Fig. S-1d). We use Δ{G} to determine automatically the number of layers in each optical image as shown in Fig. S-1(d).

2. Modeling {G} for graphene on SiO_2/Si

To model {G} of graphene layers we consider perpendicular incident light where the magnetic field is polarized in the z-direction. Our system consists of three different layers, namely, graphene, SiO_2, and Si. In each layer the magnetic field can be written as the sum of a forwards and backwards propagating wave (see Fig. S-2). The amplitude of the incident wave is set to be unity. The magnetic field can be expressed as (Wang, Y. Y.; Ni, Z. H.; Shen, Z. X.; Wang, H. M.; Wu, Y. H. Interference enhancement of Raman signal of graphene. *Appl. Phys. Lett.* **2008**, *92*, 043121):

$$H_{0z}(y)=e^{-ik_{0y}x}+Ae^{+ik_{0y}x} \quad (1)$$
$$H_{1z}(y)=Be^{-ik_{1y}x}+Ce^{+ik_{1y}x} \quad (2)$$
$$H_{2z}(y)=De^{-ik_{2y}x}+Fe^{+ik_{2y}x} \quad (3)$$
$$H_{3z}(y)=Ge^{-ik_{3y}x} \quad (4)$$

where, A, B, C, D, F, and G are parameters, H_{iz} is the magnetic field in the z-direction and K_{iy} is the wavevector in the *i-th* layer, which can be calculated by:

$$K_{iy}=\frac{2\pi}{\lambda_0}n_i \quad (5)$$

where n_i are the reflective indices, and λ_0 is the wavelength of the incident light (532 nm).
The boundary conditions are:

$$H_{jz}(-dj)=H_{(j+1)z}(-dj) \quad (6)$$

$$\frac{1}{\varepsilon_j}\frac{dH_{jz}}{dy}\Big|_{-dj}=\frac{1}{\varepsilon_{j+1}}\frac{dH_{j+1z}}{dy}\Big|_{-dj} \quad (7)$$

The thickness of graphene d_1 is estimated as $d_1=m\times 0.335$ nm, where m is the number of layers. The thickness of SiO_2 is d_2 and the Si substrate is considered as semi-infinite: H_{jz} therefore represents the magnetic field in the z-direction, $-d_j$ is the *j-th* interface and ε_j is the dielectric function in the *j-th* layer. For this calculation the following refractive indices are used: n_0=1, n_1=2.6+1.3i, n_2=1.46, and n_3=4.15+0.044i for air, graphite, SiO_2, and Si at 532 nm, respectively.

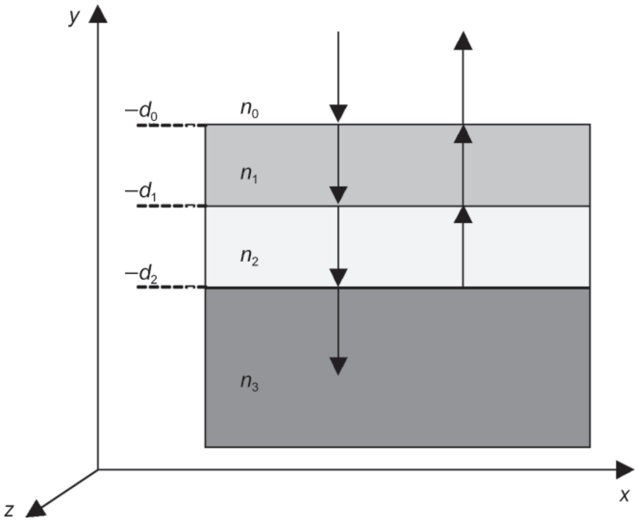


Figure S-2 Schematic illustration of light reflection and transmission in a three layer system

The two boundary conditions together with the three interfaces result in six equations with six variables A ,B, C, D, F, and G and these equations are used to determine the six variables. A is the reflectivity and is proportional to the {G} value in our optical images. To get the real {G} value of our experiment we have to multiply the reflectivity by the {G} value of the bare SiO_2/Si substrate, which is proportional to the illumination intensity used in the microscope settings. Now we can calculate how this value changes as a function of the number of graphene layers and the illumination conditions.

In Fig. S-3(a), we plot Δ{G} as a function of the number of layers (1–3 LG) and the illumination (the {G} value of the bare SiO_2/Si substrate at the same microscope illumination). Points are experimental values and lines are calculated values derived from above equations. Changes in the illumination intensities can be monitored by the {G} value of the bare SiO_2 background. The plot in Fig. S-3(a) is a plot of Δ{G} for 1-, 2- and 3-LG vs the {G} value of the bare SiO_2 next to them (proportional to the illumination). The effect of illumination was considered since on the same optical image, the illumination can change depending on the pixel position (higher illumination at the center with respect to the corners). The same

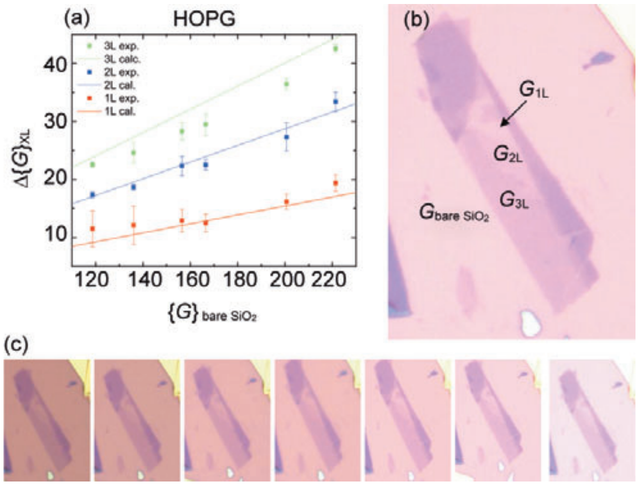
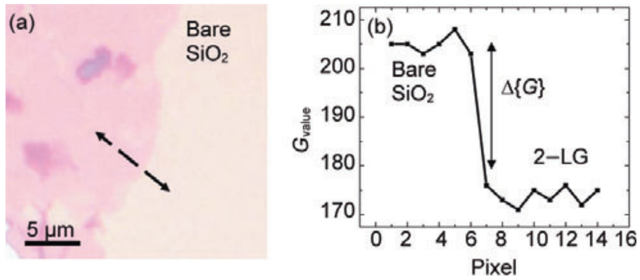


Figure S-3 (a) Empirical dependence of ΔG_{xL} on $G_{bare\ SiO_2}$ for 1-, 2- and 3-LG. Calculated results are shown as solid lines; (b) optical image of the regions used to acquire the data for (a); (c) examples of different illumination intensities which were used to extract the data plotted in (a)



$G_{bare\ SiO2}$=204.83 (illumination)

$\Delta\{G\}$= 30.95 (measured above)
Assignment: 2L
Expected for 2L = 32.23 (from Fig. S-3 (a))

Figure S-4 (a) Optical image of another CVD graphene region on SiO_2/Si; (b) {G} values corresponding to the dashed line shown in (a). $\Delta\{G\}$ values for these regions are extracted from (b) and compared to the expected values for 1–2 LG as suggested by the fits in Fig. S-3(a)

color scale settings are used for every image in the software utilized for acquiring them.

3. Quantification of the area covered by one and two CVD graphene layers ($\theta_{1\text{–}2LG}$) using optical images

For illustration, Fig. 4(a) in the main text and Fig. S-4 show the determination of $\Delta\{G\}$ for a specific CVD graphene region with a particular illumination. The observed values of $\Delta\{G\}$ closely match the expected values for HOPG-derived 1–2 LG at the same illumination. These assignments are also consistent with AFM height measurements.

To quantify $\theta_{1\text{–}2LG}$ for large areas, we use optical images taken at 50× magnification (field of view of 229 μm × 289 μm). The pixel to pixel distance is ~500 nm. The distance between thickness variations in the regions composed of 1–2 LG is usually much larger than this spatial resolution (typically on the order of a few microns). Therefore, images at this magnification and resolution are suitable for analyzing our films. Although lower magnification images could enable the quantification of $\theta_{1\text{–}2LG}$ across larger areas of the films on SiO_2/Si, they were not used due to the increase in pixel to pixel distance.

Identification of 1- and 2-LG was done in the following way. The {G} component of the optical images of clean SiO_2/Si was used as a background which is subtracted from the {G} component of the optical images of CVD graphene on SiO_2/Si. The background and CVD graphene images were taken at the same magnification and illumination conditions. The $\Delta\{G\}$ values obtained at each pixel by the subtraction were compared with the $\Delta\{G\}$ values expected for 1- or 2-LG (shown in Fig. S-3(a)) in order to label each pixel as 1- or 2-LG. This procedure was implemented with MATLAB and applied to multiple optical images in order to calculate the fraction of pixels corresponding to 1- and 2-LG in each image ($\theta_{1\text{–}2LG}$). Figure S-5 shows an example of the identification process. Notice that the pink regions of the graphene film in Fig. S-5(a) (1–2 LG) are identified effectively and tagged by the algorithm (Fig. S-5(b)).

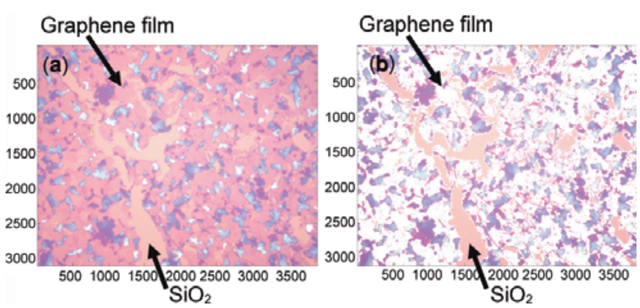


Figure S-5 (a) An optical image of a graphene film on 300 nm SiO_2; (b) the same optical image as (a) with pink regions in (a) (1–2 LG) tagged with white in (b). The film was broken (shown by the upward arrow) in order to expose part of the bare SiO_2 substrate and to test the algorithm. Images are 290 μm × 230 μm