Measuring the Thickness of Few-Layer Graphene by Laser Scanning Microscopy

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***Abstract*—This paper demonstrates laser scanning microscopy (LSM) as a viable means for quantitative and accurate measurement of reflectivity of microscopic objects/samples with diffraction-limited spatial resolution. As an example, the LSM technique along with the quantized nature of reflectivity in graphene are utilized to identify and count the number of layers of graphene few-layer flakes based on their thickness-dependent reflectivity contrast. The physical mechanism of LSM reflectivity and the associated data analysis techniques are concisely presented.**

***Index Terms*—Laser Scanning Microscopy, Optical Reflectivity Contrast, Few-Layer Graphene, Fresnel Reflection, Diffraction Limited Resolution**

Laser scanning microscopy (LSM) is a well-established technique for high-quality imaging especially in biological imaging and tomography where diffraction-limited spatial resolution is needed. Nevertheless, LSM can also be used for a variety of other applications such as characterizing local opto-electronic and microwave-photonic properties of advanced materials. Unlike a conventional microscope wherein the image is constructed at once based on the reflection and scattering of light off the sample and its propagation throughout the imaging components, LSM involves constructing the image pixel by pixel by raster-scanning a diffraction-limited laser spot over the sample. Consequently, it is only the contribution of the area under laser illumination which forms the corresponding pixel, and there is theoretically no interference effects from adjacent pixels/areas. This fact enables precise measurement of local optical/electrical properties of advanced materials and devices, for example by monitoring the perturbations in their optoelectronic or microwave-photonic response as a result of the interaction of light at the position of the laser spot with the sample. This technique has proved to be effective in studying the local RF/microwave properties of superconductors through measuring their photoresponse at low temperatures [1].

In this paper we concentrate on the reflectivity mode of LSM, where the goal is to precisely measure the reflectivity of each point on the sample under monochromatic illumination. Besides imaging the topography of the sample, local reflectivity information may be used as a non-contact and non-invasive means for identifying the material under study. For instance, we will show how LSM can differentiate different few-layer graphene materials based on their thickness-dependent reflectivity contrast. Graphene is a one-atom-thick sheet of carbon atoms ordered in a honeycomb array and has a plethora of exotic electronic and optical properties which are significant both for condensed matter physics as well as for future device applications [2]. Different graphene flakes, such as mono-layer, bi-layers, tri-layers, and so forth, drastically differ from one another with respect to their physical properties; thus, development of an efficient and precise method for their identification is of practical importance for science and industry.

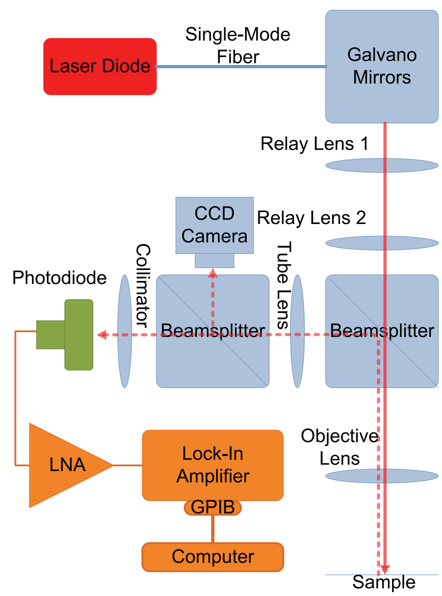

Fig. 1: The schematic of the Laser Scanning Microscopy setup used for measuring the reflectivity of the sample. A laser beam is collimated and focused into a diffraction-limit spot over the sample surface; the reflected beam, as shown by the dashed line, is then directed toward a photodiode, which generates a photocurrent proportional to the reflectivity of the area under the laser spot. The laser spot is raster scanned in the plane of the sample, by means of a pair of galvanomirrors, in order to image the sample's reflectivity.

Exfoliation of graphene from bulk graphite on Si/SiO_2 substrates is a simple and inexpensive way of preparing high-quality graphene and, therefore, is very popular.

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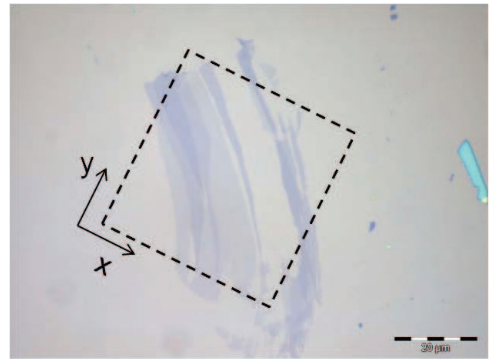
Based on the Fresnel theory of reflection, the reflectivity of Si/SiO_2/Graphene multilayer system varies with the thickness of SiO_2 and graphene [3], [4]. While the former is rather trivial, the latter provides the physical foundation for identifying the thickness of the graphene layer, and thereby the number of layers, solely based on measuring the reflectivity of the sample, which can be accurately performed by LSM.

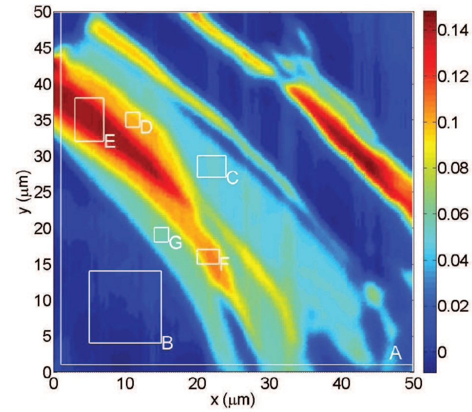
As Figure 1 illustrates, a laser diode provides the monochromatic light necessary for the LSM. The laser beam is first collimated and then focused at the surface of the sample by means of a set of standard optical lenses. A pair of galvanomirrors are used to raster scan the laser spot over the sample. The reflected light is directed to a Silicon photodiode, using a beam splitter, which converts the received light into a photocurrent that is used to construct the reflectivity image of the sample. Figure 2a shows the optical image of a mechanically exfoliated graphene flake under an ordinary optical microscope, and Figure 2b exhibits its reflectivity contrast obtained by the method described earlier. Instead of showing the reflectivity itself, we have normalized the data to present them in terms of the reflectivity contrast $C(\mathbf{r}) = [R_0 - R(\mathbf{r})]/R_0$, where R_0 is the reflectivity of a reference point on the bare substrate and $R(\mathbf{r})$ is the reflectivity of an arbitrary point at a location $\mathbf{r}$. Given the small contrast between two different graphene few-layers, the contrast image is more illustrative than a raw reflectivity image. Furthermore, it can serve as a calibration method for the experiment from one sample to the other.

Several contours, A to G, are identified at different parts of the image as examples. Figure 2c shows the histogram of the relative reflectivity, $R(\mathbf{r})/R_0$, over each of these contours all of which perfectly align with a theoretically predicted reflectivity line for few-layer graphene, as shown by vertical dashed lines for zero to ten layers, on top of the Si/SiO_2 multilayer. Atomic Force Microscopy (AFM) and Raman Spectroscopy, two widely accepted methods for identifying the thickness of graphene multilayers, have confirmed the LSM results.

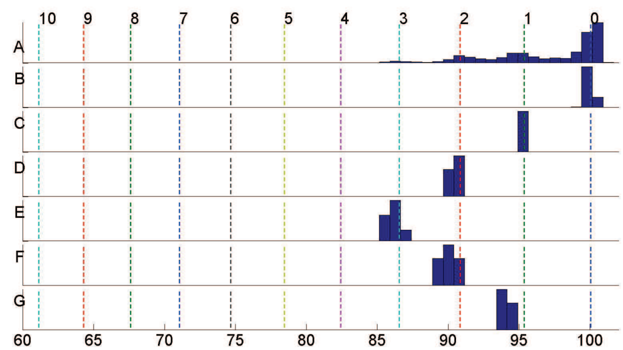
It has been shown that optical absorption in graphene is quantized and is defined only in terms of the fine structure constant $e^2/\hbar c$. Moreover, reflectivity directly relates to optical conductivity, which for an ideal Dirac Fermion system must be constant and dispersionless. [5] Therefore, the technique presented in this paper not only serves as an effective tool for identification of different graphene flakes, but also may be used to measure and relate to fundamental physical constants as well as to experimentally determine important physical quantities such as optical conductivity, complex refractive index, and their dispersion in graphene.



(a)



(b)



(c)

Fig. 2: (a) Image of the exfoliated graphene flake under an ordinary optical microscope. (b) Reflectivity contrast image obtained by the Laser Scanning Microscope. (c) The histogram of relative reflectivity ($R(\mathbf{r})/R_0$) over the contours A-G as defined in the contrast image of part (b). The zero line represents the bare substrate and by definition must equal 100% relative reflectivity.

ACKNOWLEDGMENT

This work is, in part, supported by the DOE grant number DESC0004950 and ONR/AppEl, Task D10, through grant number N000140911190.

REFERENCES

[1] A. P. Zhuravel et al., " Laser Scannign Microscopy of HTS Films and Devices", *Low Temp. Phys*, vol. 2, no. 6, 592-607, 2006.

[2] A. K. Geim and K. S. Novoselov,"The Rise of Graphene", *Nature Materials*, vol. 6, pp. 183-191, 2007

[3] P. Blake et al., "Making Graphene Visible," *Appl. Phys. Lett.*, vol. 91, 063124, 2007.

[4] P. E. Gaskel et al., "Counting graphene layers on galss via optical reflection microscopy", *Appl. Phys. Lett.*, vol. 94, 143101, 2009.

[5] R. R. Nair et al., "Fine Structure Constant Defines Visual Transparency of Graphene", *Science*, vol. 320, pp. 1308, 2008.