High resolution imaging of few-layer graphene

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In this work, we successfully demonstrate how imaging ellipsometry can be applied to obtain high-resolution thickness maps of few-layer graphene (FLG) samples, with the results being thoroughly validated in a comparative study using several complementary techniques: Optical reflection microscopy (ORM), atomic force microscopy (AFM), and scanning confocal Raman microscopy. The thickness map, revealing distinct terraces separated by steps corresponding to mono- and bilayers of graphene, is extracted from a pixel-to-pixel fitting of ellipsometric spectra using optical constants \((n = 2.7\) and \(k = 1.2\)) derived by fitting slab model calculations to averaged \(\Psi\) and \(\Delta\) spectra collected in large homogenous sample areas. An analysis of reflection spectra and contrast images acquired by ORM confirm the results by quantifying the number of graphene layers and retrieving the FLG optical constants using a simple Fresnel-law-based slab model. The morphology results are further corroborated with AFM and Raman images, the latter unambiguously verifying that the thinnest part of the FLG consists of a graphene bilayer and providing additional information of electronic origin that might help identifying subtle FLG features, such as the presence of impurities, variations in stacking order, or rolling and folding at the FLG edges. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.3694660]

INTRODUCTION

Graphene being a mono atomic thick two-dimensional crystalline solid has since its first controlled realization by micro-mechanical cleavage¹ been the subject of intense scientific interest due to its remarkable electronic, mechanical, and chemical properties.²,4–6 The main focus of the research efforts has initially been on fundamental studies and the applicability of graphene in nanoelectronics,⁴–⁶ however, an increasing attention is now also devoted to the potential applications of single and few layer graphene (FLG) and chemically modified graphene sheets in nanoscale photonics, optoelectronics, and plasmonics.⁷–¹² In the quest to realize the promising prospects in practical devices, two critical challenges are now being addressed by the research community: one being the development of fabrication techniques amenable for large scale production that provides a large area and quantity of graphene with a purity and defect density comparable to that of mechanically exfoliated graphene. Some of the approaches include growth by chemical vapor deposition (CVD) on the surface of monocristalline transition metals, segregation by heat treatment of carbon-containing substrates, and liquid phase exfoliation.⁷,¹² Secondly, the development and optimization of high throughput and non-destructive characterization techniques monitoring the relevant physical properties of the graphene layers must be available. In the context of graphene-based nanoscale devices, an investigation of the morphology and an accurate determination of the FLG thickness (number of graphene layers) will be mandatory, and several techniques have been employed to provide this information: AFM,¹³–¹⁶ scanning tunneling microscopy (STM),¹⁷ scanning (transmission) electron microscopy (S(T)EM),¹⁸,¹⁹ low-energy electron diffraction (LEED),²⁰ angle-resolved ultraviolet photoemission spectroscopy (ARUPS),²⁰ Auger electron spectroscopy (AES),²¹ Raman spectroscopy,¹³,²²–²⁶ and ORM techniques.²⁷–³³

The scanning probe microscopy techniques (AFM and STM) allow height and morphology investigations with nanometer resolution, the major disadvantages being the low throughput when considering large-area sample examination and the cumbersome interpretation of the images due to influence of tip-sample interaction¹⁴,¹⁶ and substrate topology.¹⁵,¹⁷ The electron probe-based techniques, S(T)EM, LEED, ARUPS, and AES, are also limited in throughput due to their rather complex experimental setup requiring vacuum and challenging quantitative data interpretation.²⁰,²¹ The optical techniques based on either elastically (reflected) scattered light or inelastically (Raman) scattered light can be operated at ambient conditions and have the potential of high throughput. Raman spectroscopy is the most commonly used technique capable of performing an unambiguous distinction between monolayers, bilayers and multilayers of graphene.¹³,²²,²⁴,²⁶ However, it should be noted that most Raman spectroscopy studies of graphene have been carried out on SiO₂/Si substrates as the Raman spectra of graphene on substrates like GaAs, SiC, Cu, Ni, and Ru are either complex due to the influence of the substrate or not detectable at...
all (see Ref. 21, and references therein). Similarly, in the very widely used optical contrast methods in ORM the visibility of graphene is dependent on the application of specially optimized Fabry-Pérot type SiO$_2$/Si substrates. In these investigations, the measurements of contrast images and reflection spectra have to be compared with Fresnel-law-based model calculations involving the optical constants (refractive index, $n$, and the extinction coefficient, $k$) of the FLG and substrate, in order to provide quantitative information about the number of graphene layers.

Ellipsometry is a related and commonly used optical characterization method for measuring the optical properties and thicknesses of thin films, in principle, without any specific requirements to the supporting substrate. Recently, the application of ellipsometry has been extended to FLG (Refs. 34–37) and graphene oxide layers. Kravets et al. and Weber et al. used spectroscopic ellipsometry on exfoliated single layer graphene on SiO$_2$/Si and extracted the optical properties of graphene in the visible wavelength range employing two different optical fitting models, whereas Nelson et al. applied the same technique to characterize CVD graphene, which is grown on copper foils and transferred to glass substrates. These reported spectroscopic ellipsometry measurements are conducted with relatively large spot sizes (>30 μm in diameter), which limit the lateral resolution considerably. By contrast, imaging ellipsometry can provide maps of the ellipsometric parameters with a lateral resolution below 1 μm, but information about FLG thickness has not been extracted so far.

In the present work, we combine spectroscopic ellipsometry measurements, ellipsometry imaging and Fresnel-law-based slab model calculations to obtain a high-resolution height map of the FLG sample. The presented results are compared and correlated with findings obtained by complementary graphene characterization techniques, i.e., ORM, AFM, and Raman spectroscopy.

**EXPERIMENTAL**

FLG samples prepared by micromechanical exfoliation of natural graphite are supported by a highly p-doped Si substrate with a thermally grown 90 nm thick SiO$_2$ top layer, which prior to the FLG deposition was exposed to a short dip etch in hydrofluoric (HF) acid and oxygen plasma treatment. The FLG flake to be the object of the comparative studies is supported by a highly p-doped Si substrate with a thermally grown 90 nm thick SiO$_2$ top layer, which prior to the FLG deposition was exposed to a short dip etch in hydrofluoric (HF) acid and oxygen plasma treatment.

Ellipsometry measurements were performed using an EP3se spectroscopic imaging ellipsometer (Accurion, Göttingen, Germany) under ambient conditions at room temperature. The instrument is operated as a standard null-ellipsometer with the following components in the light path: Polarizer, compensator, sample, objective, analyzer, and camera, i.e., a setup equivalent to a conventional PCSA-ellipsometer, but using focusing and camera for detection. The setup enables measurement and lateral mapping of the ellipsometry angles, $\Psi$ and $\Delta$, describing the ratio of the reflection coefficients across the sample surface: $\tan(\Psi) \exp(i\Delta) = r_p/r_s$, with $r_p$ and $r_s$ being the reflection coefficients for the light of $p$ and $s$ polarization, respectively. Overall focused images under oblique conditions are obtained with focus scanning and subsequent image reconstruction. Regions of interest (ROIs) on the sample can be selected for local ellipsometry measurements averaged over all pixels in the ROI. The light source is a 658 nm laser (used for mapping) or a Xenon lamp with filter monochromator providing 44 wavelengths (bandwidths of ±6 to ±20 nm) in the wavelength range 360–1000 nm for spectroscopic ellipsometry of selected ROIs. The reflected light is collected with a 20×(NA = 0.35) or a 50×(NA = 0.45) objective, the latter enabling a lateral resolution of approximately 1 μm. In our experiments the angle of incidence (AOI) was fixed at 42°. Analysis and modeling of ellipsometry data were performed with the software EP4Model (1.0.1) (Accurion). The software performs modeling of the ellipsometry data ($\Psi, \Delta$) by calculating Fresnel coefficients for a slab model of the sample using tabulated or measured data for the optical functions of the substrate. Typical fitting parameters in the model are thickness ($t$), refractive index ($n$), and extinction coefficient ($k$) of the FLG top layer.

The spectroscopic reflection analysis was performed on a BX51 microscope (Olympus) equipped with a halogen light source and a grating spectrometer QE65000 (Ocean Optics) with a wavelength resolution of 1.6 nm. The reflected light was collected in backscattering configuration using MP3PlanFL (Olympus) objectives with magnifications 100×(NA = 0.9) and 50×(NA = 0.75). The image area analyzed by the spectrometer is limited by a pinhole with a diameter of 150 μm, which is equivalent to a circular probing area with a diameter of 3 μm when using a 50× objective. The ORM images (1024 × 768 pixels) were captured with a black and white digital DMK31AF03 camera (The Imaging Source) with a linear responsivity verified by measurements employing neutral density filters. Contrast spectra for different numbers of graphene layers are extracted from the optical reflection measurements by calculating the wavelength-dependent contrast: $C(\lambda) = I_{FGL}(\lambda)/I_S(\lambda)$, where $I_{FGL}$ and $I_S$ are the measured optical light intensities reflected from the FLG flake and substrate surface, respectively, at the specific wavelength, $\lambda$.

AFM (Veeco, Dimension 3100) measurements using standard silicon (Si) tips in contact mode were carried out on the FLG sample in order to determine the height differences of specific terraces on the flake. The presented height profiles are averaged over several parallel line scans (windows indicated in the AFM image) for the purpose of minimizing the influence of the surface roughness on the step height estimations.

High-resolution confocal scanning Raman spectroscopy and microscopy measurements were acquired using an Alpha 300R (Witec). A 100×(0.90 NA) objective focused the linearly polarized excitation laser light ($\lambda = 532$ nm) onto the sample surface and collected all polarizations of the Raman scattered light, which was spectrally resolved employing a 600 lines/mm diffraction grating. The applied scan parameters (laser intensity, integration time, and number of points) were optimized to facilitate the best resolved images of the...
structural features on the samples with a maximum lateral resolution of ~350 nm without inflicting any heating effects, which can alter the structural or electronic properties of the FLG sample. Raman images are constructed from Raman spectra collected in every pixel in a 22 × 65 matrix with an inter-pixel distance of 150 nm, which allows for selecting the characteristic spectroscopic line to be spatially resolved.

RESULTS AND DISCUSSION

The selected FLG sample has the overall dimensions of approximately 160 µm × 100 µm and possesses a peninsula-shaped part of approximately 20 µm × 12 µm (within the dotted square in Fig. 1(a)). The contrast variations in the ellipsometric (Fig. 1(a)) and optical reflection (Fig. 3(b)) image indicate four terraces with increasing height on the peninsula and two areas of different thickness on the FLG ‘mainland’ (ROI1 and ROI2 in Fig. 1(a) and upper left corner in Fig. 2(b)).

The actual structure under investigation is a four layer slab structure comprised of crystalline Si substrate, SiO₂ layer, FLG top layer and air (Fig. 1(b)). It should be noted that we will consider the FGL to be optically isotropic, as the ellipsometry measurement is predominantly sensitive to the in-plane optical response of the graphene layer, given that the optical path along the anisotropy axis (c-axis) is restricted to a few graphene layers.37 In order to determine the optical constants and thickness of the FLG, it is imperative to know or determine the optical properties of the underlying layers, i.e., their optical functions and thicknesses.

The optical functions of crystalline Si are well known (and implemented in the EP4Model simulation software), and the thickness of the substrate is considered semi-infinite in the model calculations. As for SiO₂, the extinction coefficient is negligible and the refractive index is largely dispersion-free, though, dependent on the quality and stoichiometry of the amorphous layer. Since the SiO₂ layer in the present case is formed by a well-controlled thermal growth process, we choose to use the tabulated SiO₂ optical functions implemented in the EP4Model and determine the SiO₂ layer thickness by numerical inversion of Ψ and Δ spectra collected on the bare SiO₂ surface in the vicinity of the FLG flake (ROI0 in Fig. 1(a)). From the measured Ψ and Δ data and the corresponding fitted curves obtained from the model calculations (triangle symbols and blue curve in Figs. 1(c) and 1(d)), a SiO₂ layer thickness of 87.5 nm ±0.1 nm is derived (table in Fig. 1). Knowing this value, the optical constants (n, k) and thickness (t) of the FLG in ROI1 and ROI2 are subsequently determined by fitting the full four layer slab model to Ψ and Δ spectra collected from the respective ROIs (Figs. 1(a), 1(c), 1(d)). The obtained results are n = 2.68 ± 0.18, k = 1.24 ± 0.10, and t = 3.0 ± 0.1 nm for ROI1 and n = 2.65 ± 0.17, k = 1.21 ± 0.10, and t = 2.8 ± 0.1 nm for ROI2 (table in Fig. 1). With a single layer graphene thickness being 0.335 nm, the measured thicknesses of 2.8 nm and 3.0 nm correspond to 8–9 layers of graphene on the FLG mainland, and the contrast in the ellipsometry image suggests a descending number of layers on the peninsula part. The experimental values for the optical constants are in good agreement with reported results obtained on graphite by

![Fig. 1.](image-url)
optical reflection measurements ($n = 2.67, k = 1.3$ at $\lambda = 532$ nm and $n = 2.73, k = 1.42$ at $\lambda = 633$ nm)\textsuperscript{41} and by spectroscopic ellipsometry ($n = 2.52, k = 1.94$ at $\lambda = 546$ nm),\textsuperscript{43} and also comparable to results recently obtained on graphene by spectroscopic ellipsometry, where Kravets et al.\textsuperscript{34} reported $n = 2.7–2.8$ and $k = 1.4–1.6$ and Weber et al.\textsuperscript{35} $n = 2.5–2.7$ and $k = 1.1–1.4$ in the wavelength range 350 nm to 750 nm. For a detailed discussion on the optical functions of graphite, FLG, and graphene including the significant discrepancies in the reported values, we refer to the literature.\textsuperscript{32,42,43}

Assuming that the optical properties of Si and SiO$_2$ are spatially homogenous and that the optical functions of graphene and FLG only differ slightly,\textsuperscript{29} the layer thickness is the only fitting parameter, and it is possible to convert a recorded map of $\Psi$ or $\Delta$ data into a thickness map of the FLG sample. Thus, we have performed a pixel-to-pixel fitting of a measured $\Delta$-map recorded at an illumination wavelength of $\lambda = 658$ nm using the slab model (Fig. 1(b)), the tabulated dispersion functions for SiO$_2$ and Si, and an average value of the measured complex refractive index for FLG ($n = 2.7, k = 1.2$). The black and white spot in (a) and (b), respectively, are due to a very thick graphite piece exceeding the limit of the height scale (saturation). Averaged height profiles are plotted for (c) the multi-layer steps and (d) the single layer step averaged in the indicated windows in (a).

FIG. 2. (Color online) (a) Thickness map and (b) the corresponding spatial 3 D perspective view of the FLG sample obtained from pixel-by-pixel fitting of a measured $\Delta$-map recorded at an illumination wavelength of 658 nm. The fitting is based on the slab model (Fig. 1(b)), tabulated dispersion functions for SiO$_2$ and Si, and an average value of the measured complex refractive index for FLG ($n = 2.7, k = 1.2$). The black and white spot in (a) and (b), respectively, are due to a very thick graphite piece exceeding the limit of the height scale (saturation). Averaged height profiles are plotted for (c) the multi-layer steps and (d) the single layer step averaged in the indicated windows in (a).

double graphene steps (0.67 nm) with the exception that the first step from the substrate surface to the lowest terrace (~0.9 nm) is slightly larger than a double layer. The latter observation is not unexpected, as several groups have reported highly varying AFM thickness measurements of single graphene layers on SiO$_2$, ranging from 0.35 nm to 1.6 nm.\textsuperscript{1,14,16} These variations have been attributed to the difference in tip-substrate and tip-graphene interaction, to trapped water between the SiO$_2$ and FLG, to the roughness of the SiO$_2$ surface, to adsorbed molecules on the SiO$_2$ and FLG surfaces, and to interactions between the FLG and the supporting substrate.\textsuperscript{1,14,16,44} It is highly probable that the increased thickness of the lowest terrace in our sample is due to trapped adsorbates such as water. Although Kravets et al. in their fitting model, when extracting the optical functions of graphene from ellipsometry measurements, introduce a thin Cauchy sublayer representing a spacer of water and air under the graphene,\textsuperscript{34} we have omitted this extra layer in the model to avoid introducing more fitting parameters that may not be determined accurately from the data. The simple model also facilitates a comparison of results between our measurements using different characterization techniques. Furthermore, we observe an offset of 2–3 Å between the thickness values
obtained from modeling of ellipsometry data will always be influenced by any characteristics of a sample that deviates from the ideal slab model (Fig. 1(b)). A relevant example (noticeable in Fig. 2(a), Fig. 2(b), and especially in Fig. 3(b)) is the sample edge features that can be attributed to rolling or back-folding of the graphene layers on themselves. The ellipsometric (and also the Raman) spectra collected in these areas will be affected if the ordering and the bonding of the back-folded layer to the original surface deviate from the crystalline properties of the pristine FLG “bulk” layers. Consequently, the thickness values obtained by model-based numerical inversion of the ellipsometric spectra, should be interpreted cautiously in samples areas containing these edge features.

Using ORM with a 50×(NA = 0.75) objective lens, we have measured contrast spectra \( C(\lambda) \) (Fig. 3(a)) on each of the specific terraces as indicated by the corresponding symbols in the contrast image (Fig. 3(b)), which has been recorded at the wavelength of optimum contrast \( \lambda = 550 \text{ nm} \) ± 10 nm. The arrows point out distinct step features that are also identifiable in the AFM and Raman images presented later. We are able to obtain supplementary quantitative information about the thickness and optical constants of the FLG sample by fitting calculated contrast spectra (solid curves) to the experimental data (symbols) presented in Fig. 3(a). The contrast spectra are calculated by applying the matrix method when determining the reflection coefficient of the multilayered slab model structure (Fig. 1(b)). In setting up the matrix of the individual layers, we take into account the angle of light incidence, the optical functions of the material, and the thickness of the respective layer. Consistently, we use the same tabulated optical functions of Si and SiO\(_2\) as well as thickness of SiO\(_2\) \( t = 87.5 \text{ nm} \) employed in the ellipsometry modeling, whereas the thickness of the FLG layer is increased in integer multiples of 0.335 nm (thickness of single graphene layer). The reflection coefficients of the slab model with and without FLG and hence also the contrast spectra are computed by integrating the optical response of the slab model over all incident angles \( \text{NA} = 0.75 \) and polarizations of light using a light intensity distribution described by a Gaussian beam profile. This approach predominantly determines the in-plane optical response of FLG, since the out-of-plane compared to the in-plane response is much smaller and further diminished in the weighted integration over incidence angles given by the NA. Using the implication that the FLG optical properties are independent of thickness, and in addition, assuming dispersion-free optical constants, the calculated contrast spectra (solid curves, Fig. 3(a)) can be fitted to the experimental values (symbols, Fig. 3(a)), where an excellent agreement is achieved for FLG optical constants of \( n = 2.87 \) and \( k = 1.24 \), respectively 8% and 2% higher than the values obtained by our ellipsometry investigations. The experimental contrast values for the second terrace (Fig. 3, blue dots) are almost consistently slightly higher than the contrast curve calculated for a thickness of four layers of graphene (N = 4). The data are dominantly determined by the thickness of this terrace, but will also have minor weighted contributions from the neighboring terraces, as the probing spot diameter is 3 \( \mu \text{m} \) and thereby exceeding the width (~2 \( \mu \text{m} \)) of the second terrace. In the present measurements the probing area

obtained in ROI1 and ROI2, when comparing the spectroscopic results listed in the table of Fig. 1 \( (t = 2.8 \text{ nm and } t = 3.0 \text{ nm, respectively}) \) and the single-wavelength mapping values from the height profile in Fig. 2(d) \( (t = 2.5 \text{ nm and } t = 2.8 \text{ nm, respectively}) \). This illustrates the minor numerical difference between thicknesses obtained from a spectroscopic data set averaged over a region-of-interest and mapped thicknesses obtained at a fixed wavelength in single pixels. In contrast, the difference between the thickness of what appears to be the same terrace on the peninsula (top left part of linescan in Fig. 2(c) and ROI1 (lower left part of linescan in Fig. 2(d)), we attribute to the aforementioned trapping or adsorption of molecules, actual sample inhomogeneities in the substrate layers, or variations in the FLG height due to formation of ripples or bubbles in the graphene layers. Finally, we remark that results

FIG. 3. (Color online) (a) Experimental (symbols) and calculated (solid curves) optical contrast spectra obtained on the respective terraces on the FLG sample as indicated by the corresponding symbols in the contrast image (b). The contrast spectrum indicated by the olive colored triangle is collected in ROI2 (Fig. 1(a)) outside the contrast image (b). All spectra are measured with a 50× objective and are calculated taking NA = 0.75 into account. (b) The contrast image has been recorded using a 100× objective and an optical bandpass \( (\lambda = 550 \text{ nm} \pm 10 \text{ nm}) \) filter in front of the camera, which ensures a maximum contrast between the layers (according to the contrast differences at \( \lambda = 550 \text{ nm} \) in (a)). The arrows point out distinct step and defect features, which are also identifiable in the AFM image (Fig. 4(a)) and the Raman images (Figs. 5(c)–5(e)).

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probably has a slightly larger overlap with the terrace above compared to the one below. The combined ellipsometry and ORM measurements unambiguously conclude that the thicknesses of the terraces given in numbers of graphene layers are (Figs. 2 and 3(a)): 2, 4, 6, 7, and 8 (the latter number being the thickness in ROI2).

The morphological AFM image (Fig. 4(a)) reveals the two steps separating the first three terraces on the FLG peninsula, and the step height profiles (Fig. 4(b)) averaged in the indicated windows once again confirm, that each of the step heights corresponds to a height of two layers of graphene (0.67 nm). Furthermore, we can clearly identify the two step features also present in the contrast image (gray and white arrows in Fig. 3(b)). The AFM measured height (not shown) of the first terrace with respect to the SiO2 surface is ~1.9 nm, which exceeds the expected value of the confirmed bilayer by ~1.2 nm. The sources of this discrepancy have been discussed above, and our value is within the range 0.35–1.6 nm reported by others.1,14,16 Based on our direct comparison between Ellipsometry/ORM and AFM as well as previous results, we note that thickness measurements of graphene with AFM appear to be subject to a considerable systematic error due to varying tip-sample interactions.

Raman spectroscopy measurements performed on the four terraces of the peninsula (Fig. 5(a)) reveal lines at ~975 cm⁻¹ (Si line) stemming from the supporting substrate and characteristic graphene lines at ~1580 cm⁻¹ (G line) and ~2700 cm⁻¹ (2D line). Note the characteristic rise in the intensity of the G peak and changing peak form of the 2D line as well as the increased damping of the Si line, when the thickness of the FLG increases.22–26 The absence of a very

![AFM image of the FLG sample area with three terraces and (b) height profiles of the two steps (averaged in the indicated windows) separating the terraces. The height of both steps corresponds to the thickness of two layers of graphene. The colored arrows indicate the positions of the same distinct step features found in the contrast image (Fig. 3(b)) and the Raman image (Fig. 5(c)).](image1)

![Raman spectra collected on the FLG terraces. The use of the symbols in the graphs is correlated with the use of the symbols in Fig. 3 and (c). The Raman spectra clearly display the intensity variations of the silicon line at ~975 cm⁻¹, graphene G line at ~1580 cm⁻¹ and the graphene 2D line at ~2700 cm⁻¹. (b) Optical image indicating the sample area where Raman images of the integrated intensity of (c), (d) the G line and (e) the defect line (bottom image) have been recorded with integration intervals of 1568–1605 cm⁻¹ and 1316–1376 cm⁻¹, respectively. (d) The image demonstrates that the crack defect (blue arrow) is also visible in the right part of the G line image (c) if the contrast is adjusted. The colored arrows indicate the positions of the same distinct step features found in the contrast image (Fig. 3(b)) and AFM image (Fig. 4(a)).](image2)
narrow 2D line with an intensity exceeding that of the G line is the most unambiguous evidence that the lowest terrace is not a single layer of graphene. However, based on the Raman spectra it is very difficult to extract any further direct quantitative information regarding the respective number of layers in the remaining terraces, as the G line intensity only increases linearly with the number of layers up to five layers, hereafter the signal starts asymptotically to approach the bulk value of graphite. Nevertheless, as the present FLG has a maximum of 8 stacked layers, a Raman image of the integrated intensity of the G line (1568 - 1605 cm$^{-1}$) obtained in the area marked in the optical image (Fig. 5(b)) reveals the four terraces (Fig. 5(c)), and also the crack (Fig. 5(d)) in the lowest terrace, when adjusting the contrast level. Interestingly, we also observe some additional features that are not visible in the ORM contrast image (Fig. 3(b)) or in the AFM image (Fig. 4(a)). The colored arrows correlate with the positions of step and defect features indicated by the arrows in the ORM and AFM images. From a comparison of the images it is evident that the integrated G line intensity in the vicinity of the step (gray arrow) exhibits variations that are not only due to a change in the number of stacked layers, but also variations that must be of electronic origin. In an attempt to directly image and accentuate the step features, a Raman image was recorded using the integrated intensity of the D line at $\sim$1350 cm$^{-1}$ (Fig. 5(e)). The 2D line at $\sim$2700 cm$^{-1}$, being the overtone of the D line, is always present in the Raman spectrum of pristine graphene and graphite, whereas the D line is absent. The appearance of the D line is associated with the occurrence of defects and disorder (e.g., at step edges), which affect the structure symmetry and electronic properties. Indeed we observe a pronounced D line signal originating from the crack defect in the lowest terrace (blue arrow in Fig. 5(e)), whereas the signal expected from the step edges is too low to discern the outline of the steps or any defects that might be responsible for the variation of the G line intensity in the vicinity of the step (gray arrow). The origin of these intensity variations remains unclear; however, we would like to note that also edge rolling and back-folding, contamination, impurities, and stacking order can influence the G and 2D lines in the Raman spectra of FLG.

**CONCLUSIONS**

To conclude, we have used the complementary characterization techniques imaging ellipsometry, optical reflection microscopy, AFM and scanning confocal Raman microscopy to determine the thickness variation and optical properties of FLG supported by a SiO$_2$ film on Si. In particular, we have successfully demonstrated the possibility of extracting a high resolution thickness map by performing a pixel-to-pixel fitting of ellipsometric $\Delta$ spectra recorded in every image pixel. In this fitting procedure we use a FLG refractive index ($n$) of 2.7 and extinction coefficient ($k$) of 1.2, values derived by fitting slab model calculations to experimental $\Psi$ and $\Delta$ spectra collected and averaged in specific regions of interest. The thickness map reveals distinct terraces separated by steps, which have heights corresponding to monolayers or bilayers of graphene. The findings are in excellent agreement with the optical constants ($n = 2.87$, $k = 1.24$) and thickness values obtained from ORM, where we in the framework of Fresnel coefficient calculations have fitted contrast curves to experimental contrast spectra measured on each of the FLG terraces. Also, the contact mode AFM measurements confirm the morphological results, as we are able to image three of the terraces separated by bilayer graphene step heights, and we can perform a direct spatial correlation with the thickness map and contrast image when identifying distinct step features. The intensity variations of characteristic G and 2D lines in Raman spectra recorded on each of the terraces are consistent with the results of the other characterization methods, and they unambiguously verify that the thinnest part of the FLG consists of a graphene bilayer and not a monolayer. In addition, the Raman image based on the integrated intensity of the G line displays some spatial intensity variations in the vicinity of a step edge. These features are not observed in the contrast image or in the AFM image and are probably of electronic origin.

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40. The optical functions implemented in ellipsometry EP4Model, version 1.0.1, (Accurion GmbH, Göttingen, Germany) are based on the open SOPRA n&k database (http://www.sopra-sa.com).