

Total Color Difference for Rapid and Accurate Identification of Graphene

Libo Gao, Wencai Ren,* Feng Li, and Hui-Ming Cheng*

Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, 72 Wenhua Road, Shenyang 110016, People's Republic of China

Graphene, as a one-atom-thick flat allotrope of carbon, has been recently attracting increasing interests from both fundamental physics study and potential applications. It exhibits high crystal quality and ballistic transport in a submicron scale (even under ambient conditions), and its charge carriers accurately mimic massless Dirac fermions.¹ The unusual band structure of graphene allows one to probe interesting transport phenomena such as anomalous integer quantum Hall effect.^{2,3} Combined with its nanometer scale, graphene is also suggested to be an excellent candidate for ultra-high-frequency transistors.¹ However, the properties of graphene are closely correlated with its structures. A variation of the number of graphene layers may result in striking changes of their electronic properties,¹ so rapid and accurate identification of the number of graphene layers can be a crucial step for their selective preparation of graphene and the fabrication of graphene-based devices.

Compared to atomic force microscopy (AFM),^{4,5} scanning electron microscopy (SEM),⁶ high resolution transmission electron microscopy (HRTEM),⁷ and Raman spectroscopy,^{8–11} optical methods offer the potential for rapid and nondestructive characterization of large-area graphene samples.^{6,12–14} Recently, several routes have been proposed for improving the image contrast of graphene, such as narrow band illumination and selecting appropriate substrate,^{12,14} and reflection and contrast spectroscopy was used for approximate identification of the number of graphene layers.¹³ In contrast to a monochromatic light source, the use of white light allows rapidly sorting out graphene thickness regions because graphene with

ABSTRACT For rapid and accurate identification of graphenes by optical images, a total color difference (TCD) method is proposed and demonstrated based on a combination of reflection spectrum and International Commission on Illumination color space. The preferential thickness of different dielectric films covered on a Si substrate is well elucidated, and a 72 nm thick Al_2O_3 film is found to be much better than the commonly used SiO_2 or Si_3N_4 films. The TCD both between monolayer graphene and substrate and between graphene of different layers can be further improved by appropriately narrowing the wavelength range of the light source. Moreover, the influences of the objective lens in a real-world optical system on the TCD are also discussed. These findings provide useful information for rapid evaluation of the layer range of graphenes simply by different color bands and for accurate and reliable layer identification due to large TCD values, which opens up the possibility for the nondestructive identification and physical property measurements of graphene with an optical microscope.

KEYWORDS: graphene · total color difference · number of layers · substrate · light source · objective lens · optical method

different ranges of thickness can exhibit different color bands that can be easily appreciated by the naked eye.⁶ However, only marginal color difference between monolayer graphene and substrate and between graphenes with different layers was observed even for the well-accepted optimum substrate.¹² Moreover, the selection of appropriate substrate in white light needs an effective and reliable theoretical guide, although several groups have tried to elucidate the preferential SiO_2 thickness based on monochromatic contrast calculation.^{12,14} These issues prevent the use of white light for rapid and accurate identification of graphene layers.

Here we propose a total color difference (TCD) method, based on a combination of the reflection spectrum calculation and International Commission on Illumination (CIE) color space,¹⁵ to quantitatively investigate the effect of light source and substrate on the optical imaging of graphene. The preferential substrates suitable to detect graphene are well determined and elucidated, and a 72 nm thick Al_2O_3 film is found to be much better at characterizing

*Address correspondence to cheng@imr.ac.cn, wcren@imr.ac.cn.

Received for review May 19, 2008 and accepted July 09, 2008.

Published online July 25, 2008. 10.1021/nn800307s CCC: \$40.75

© 2008 American Chemical Society

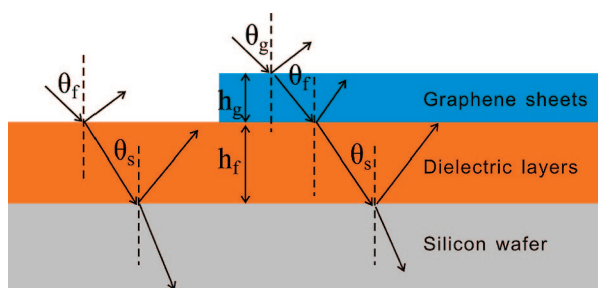


Figure 1. Optical reflection and transmission schematic for a layered thin-film system consisting of a dielectric film (Al_2O_3 , SiO_2 , or Si_3N_4) on silicon wafer (left part) and graphene added on the dielectric film (right part).

graphene than SiO_2 and Si_3N_4 films. Moreover, the image contrast of graphene layers can be remarkably improved if the wavelength range of light source is narrowed appropriately, instead of the commonly recommended monochromatic light.^{12,14} The influences of the objective lens in a real-world optical system on the TCD are also discussed.

RESULTS AND DISCUSSION

The image contrast of graphene in visible light is an integration of all the contrasts for each wavelength component. Consequently, a colorful image is obtained in this case instead of the black-and-white image for narrow band illumination, and a new standard instead of the commonly used mono-

chrome contrast is required due to the color factor. The CIE color space includes both brightness and color and is modeled on the color sensitivity and perception of human eyes.¹⁵ Therefore, we propose TCD as a color standard in this paper, based on a combination of the reflection spectrum calculation and CIE color space, to quantitatively describe the contrast of color image.

The schematic of the optical reflection and transmission for a layered thin-film substrate is shown in Figure 1. For a dielectric film on a wafer (here Si), the substrate has two interfaces; with the addition of graphene, a third interface is added. Because a portion of the beam is reflected from each interface and the rest is transmitted, a number of optical paths are possible. The amplitude of the reflected beam is a result of interference between all the optical paths and is determined by the wavelength of the incident light (λ), their incident angles (θ), refractive indices of all layers (n), and their thicknesses (h). Thus, the total reflectivity of light from the triple-layer system can be described by¹⁶

$$R(\lambda) = \left| \frac{r_1 + r_2 e^{-2i\delta_1} + r_3 e^{-2i(\delta_1 + \delta_2)} + r_1 r_2 r_3 e^{-2i\delta_2}}{1 + r_1 r_2 e^{-2i\delta_1} + r_1 r_3 e^{-2i(\delta_1 + \delta_2)} + r_2 r_3 e^{-2i\delta_2}} \right|^2 \quad (1)$$

where $r_1 = (n_0 - n_g)/(n_0 + n_g)$, $r_2 = (n_g - n_f)/(n_g + n_f)$, and $r_3 = (n_f - n_s)/(n_f + n_s)$ are the reflection coeffi-

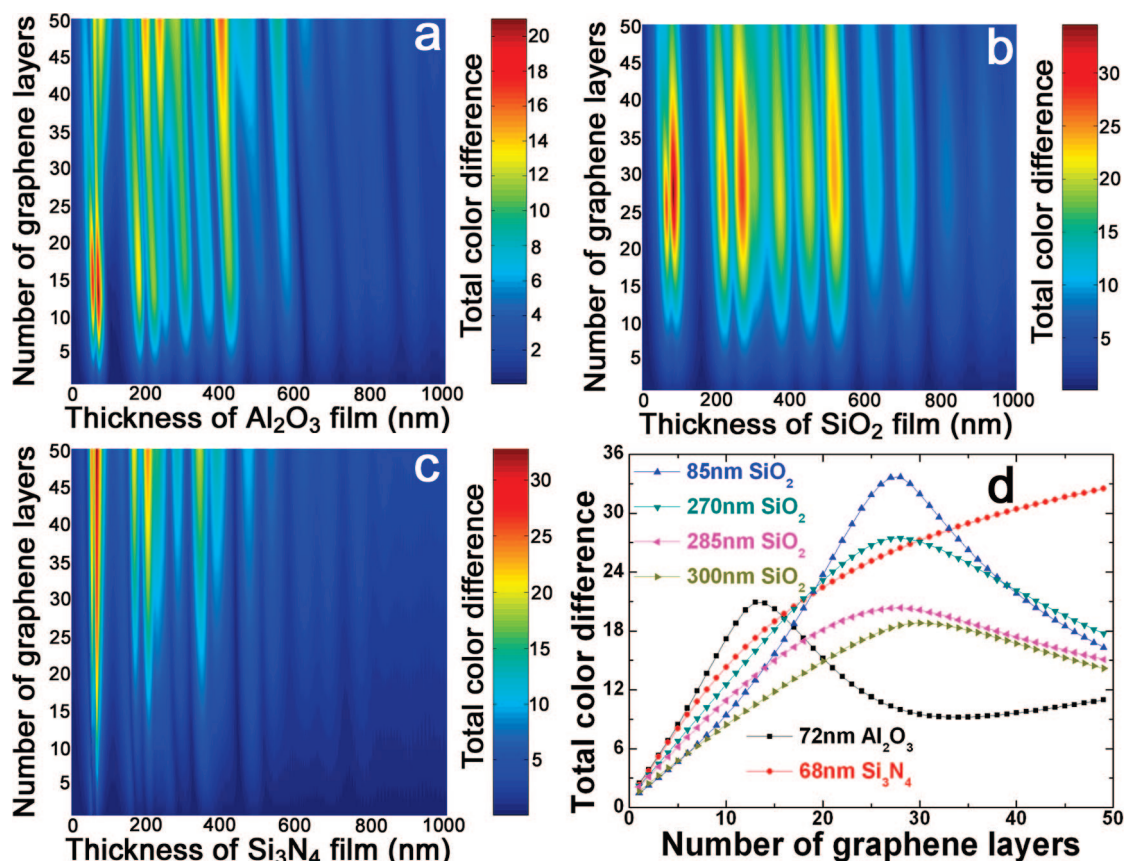


Figure 2. Contour plot of TCD as a function of the number of graphene layers and the thickness of Al_2O_3 film (a), SiO_2 film (b), and Si_3N_4 film (c). (d) Comparison of TCD for different preferential dielectric film thicknesses.

icients for different interfaces, $\delta_1 = (2\pi/\lambda)n_g h_g \cos \theta_g$ and $\delta_2 = (2\pi/\lambda)n_f h_f \cos \theta_f$ are the phase shift when the light passes through graphene and the dielectric film, which is determined by the path difference of two neighboring interfering light beams. The refractive indices of the wafer (n_s) and dielectric film (n_f , such as SiO_2 , Al_2O_3 , and Si_3N_4) are wavelength-dependent, and the refractive index of air (n_0) is 1.0003. Ni *et al.*¹³ studied the effect of refractive index of graphene on the calculated optical contrast, and they found that the calculated contrast spectra are in good agreement with the experimental results when the refractive index was adopted as 2.0–1.1*i* in the calculations. Moreover, it is worth noting that the use of this value can well reproduce the contrast spectra not only for the monolayer graphene but also for the graphene of 2–10 layers. Therefore, the refractive index of graphene (n_g) is referred as 2.0–1.1*i* in our calculations.¹³ However, we should point out that a different refractive index should be adopted instead of 2.0–1.1*i* for very thick graphene because of the increased interlayer interaction and the more complex interference behavior of interfering light from each layers. For example, the refractive index of bulk graphite is well accepted to be 2.6–1.3*i*. The incident angles at the interface of air–graphene (θ_g) and graphene–dielectric film (θ_f) also affect the image contrast, which is fixed at 0° in our calculations first. The thickness of graphene can be estimated as $h_g = 0.35 + 0.33t$ (t is the number of layers),⁸ and h_f represents the thickness of dielectric film. The reflection without graphene layers is calculated by using $h_g = 0$. Note that the total reflectivity of a triple-layer system is also related to the light wavelength since the optical constants of a material is wavelength-dependent.

To reproduce the color difference of graphene with different layers in white light, we combine the calculated total reflectivity of light with CIE color space¹⁵ for further analysis. The gamut of described colors is used as a high fidelity device-independent color system that closely approximates the perceivable range and scope of colors. Any given spectrum can be decomposed into X , Y , and Z tristimulus components by applying the CIE color-matching equation¹⁷ to the spectrum. Here, the X , Y , and Z components of the reflected light can be calculated by integrating its wavelength spectrum k times the color-matching functions, and CIE 1976 $L^*a^*b^*$ color space is also imported to obtain the color difference between sample and substrate, which is transformed from XYZ space.¹⁵ They are given as follows:

$$\begin{cases} X = k \int_{\lambda} S(\lambda)R(\lambda)x(\lambda)d\lambda \\ Y = k \int_{\lambda} S(\lambda)R(\lambda)y(\lambda)d\lambda \\ Z = k \int_{\lambda} S(\lambda)R(\lambda)z(\lambda)d\lambda \end{cases} \quad (2)$$

$$\begin{cases} L = \begin{cases} 116(Y/Y_0)^{1/3} - 16 & (Y/Y_0 > 0.008856) \\ 903.3Y/Y_0 & (Y/Y_0 \leq 0.008856) \end{cases} \\ a^* = \begin{cases} 500[(X/X_0)^{1/3} - (Y/Y_0)^{1/3}] & (X/X_0 > 0.008856) \\ 3893.5[X/X_0 - Y/Y_0] & (X/X_0 \leq 0.008856) \end{cases} \\ b^* = \begin{cases} 200[(Y/Y_0)^{1/3} - (Z/Z_0)^{1/3}] & (Z/Z_0 > 0.008856) \\ 1557.4[Y/Y_0 - Z/Z_0] & (Z/Z_0 \leq 0.008856) \end{cases} \end{cases} \quad (3)$$

$$\Delta E_{ab}^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2} \quad (4)$$

where $S(\lambda)$ is the total light source power, X_0 , Y_0 , and Z_0 are tristimulus components of CIE standard illuminance; L^* is called psychological lightness, and a^* and b^* are psychological chroma; ΔL^* is called lightness difference which represents the contrast without the effect of color factors and is similar to that presented under monochromatic light, and Δa^* and Δb^* are color difference; ΔE_{ab}^* is TCD, which is the final image contrast between graphene and substrate. According to the National Bureau of Standards (NBS), the image can be distinguished slightly if the TCD value is more than 0.5, and they become well distinguished if the value is more than 1.5. The larger the value, the easier we can identify the difference.

In our calculations, the halogen light spectrum was used to determine the TCD of graphene. If we change to another spectrum-known illumination lamp, such as a D65 lamp, the TCD values may change in a marginal way because of the slight difference between their light spectra, but the apparent color will obviously change (Figure S1 in Supporting Information). If the wavelength range of light source is narrowed, only the light spectrum in eq2 is changed in the TCD calculations.

The XYZ color parameters from eq 2 can be also converted to a device-dependent color scheme, such as red-green-blue (RGB). The conversion to RGB is accomplished by multiplying the XYZ vector by a transformation matrix as follows:¹⁷

$$\text{RGB} = M * XYZ \quad (5)$$

The transformation matrix M is generated by the monitor's phosphor chromaticity coordinates and the reference white of the light source. Therefore, the XYZ color parameters for a colorful image of graphene can be directly obtained from their RGB, and the corresponding TCD values are subsequently derived by eqs 3 and 4. These experimentally derived TCD values can be used to characterize the number of graphene layers by comparing them with those calculated TCD values for different layers of graphene on the same imaging conditions.

Figure 2a–c displays one of the main calculated results with TCD method: the contour plot of TCD as a function of number of graphene layers for different dielectric film (Al_2O_3 , SiO_2 , and Si_3N_4) thicknesses. It is clearly seen that only a few dielectric film thicknesses

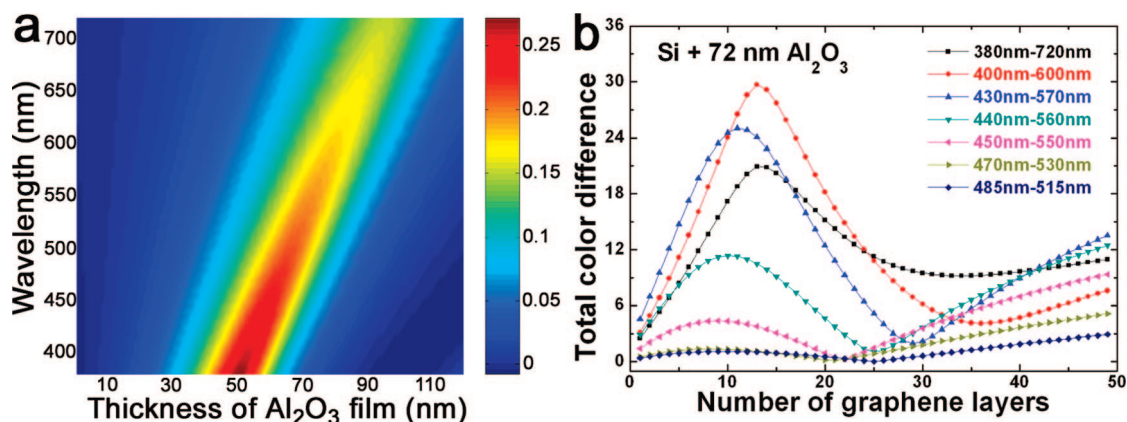


Figure 3. (a) Calculated contrast of graphene as a function of wavelength for different thicknesses of Al₂O₃ film, using the method reported by Meyer *et al.*⁷ (b) Calculated TCD of graphene located on the top of Si substrate with a 72 nm Al₂O₃ film for different wavelength ranges of light source.

are suitable for detecting graphene under white light since their corresponding TCD is larger than 1.5, a threshold for clear visibility of the image observed by the naked eye. Note that our results well reproduce the experimentally obtained preferential thicknesses of 285,¹³ 300,⁴ and 465 nm⁶ for SiO₂, and of 68 nm¹⁴ for Si₃N₄ films, for the observation of monolayer graphene. Several groups tried to explain the preference of SiO₂ thickness for high contrast in the visible range; however, all the explanations are based on the presence of maximum contrast at a specific wavelength. Actually, the real contrast of graphene in visible light is an integration of all the contrasts for each wavelength component. Therefore, it is difficult to tell which thickness of SiO₂ is the best choice to identify monolayer graphene for the white light case using the previously reported methods. The above analyses suggest that there are some limitations for prediction on the preferential dielectric film thickness only based on reflection calculation. On the contrary, our TCD method can offer reliable information to guide the selection of appropriate substrate for rapid and accurate identification of graphene using white light.

In order to quantitatively determine the suitable substrate, we list the TCD values as a function of the number of graphene layers for different preferential dielectric film thicknesses (Figure 2d). One important feature of Figure 2d is that some TCD inflexions appear with an increase of the number of graphene layers, which is suggested to correspond to the change of color band (Figure S2 in Supporting Information). This was observed before on the Si substrate with SiO₂ film,^{4,6} where graphene with many layers exhibits different color band from a few-layer one. The different color bands allow one to easily sort out the thickness range of graphene by the naked eye. These results are quite different from those obtained only using a monochromatic light source,^{12,14} where the thickness difference cannot directly be sorted out from the image contrast only by the naked eye without further analyses.

More interestingly, our calculated results also show that the inflection for the same layer range can be tuned by different substrates, which is helpful for the selection of the most suitable substrate for the rapid evaluation of the layer range of graphene. As pointed out above, large TCD both between monolayer graphene and substrate and between graphene with different layers is required for accurate characterization of graphene layers. Therefore, among the studied dielectric films, 72 nm thick Al₂O₃ film is the best for rapid and accurate optical characterization of graphene under visible light using an optical microscope (OM), due to the high sensitivity of color bands on the number of layers and larger TCD between different layers of graphene.

Narrow band light can be considered as light with a specific wavelength range; therefore, our method can also be easily used to investigate the effect of wavelength of light source on the image contrast of graphene. In our calculations, the calculated reflection result¹² shown in Figure 3a is used to get the optimized wavelength of monochromatic light for a specific Al₂O₃ thickness. The investigated wavelength ranges are then selected using the calculated wavelength as center. Figure 3b illustrates our calculated results for 72 nm thick Al₂O₃ film, and we clearly see that the TCD between graphene with one layer difference is remarkably increased when the wavelength range is decreased suitably. For example, the TCD values between monolayer graphene and substrate and between graphene with one layer difference are increased from 2.3 to 4.6 and from 1.7 to 3.1, respectively, when the wavelength range of light source is changed from 380–720 nm to 430–570 nm. Therefore, the visibility of monolayer graphene and the contrast between graphene with different layers are both dramatically improved, which is very important for the accurate determination of graphene layers. However, if the wavelength range of light source is too narrow, approaching monochromatic light, the TCD becomes smaller on the contrary. This im-

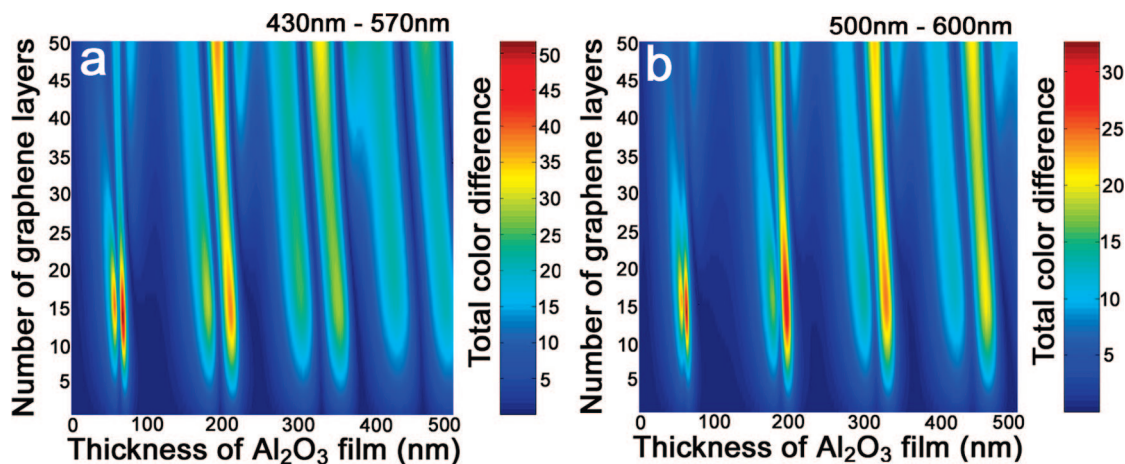


Figure 4. Calculated TCD of graphene located on the top of Si substrate with an Al_2O_3 film for a preferential wavelength range of 430–570 nm (a) and a random wavelength range of 500–600 nm (b). The optimum substrate film is 70 and 210 nm thick Al_2O_3 films for the light source of 430–570 nm, but it is 65 and 197 nm thick Al_2O_3 films for 500–600 nm light source.

plies that monochromatic light is not the best choice for the observation of graphene using the optical method. Moreover, it is worth noting that the positions of inflexions can also be tuned by changing the wavelength range, and the inflexions shift to the position of fewer layers as the wavelength range becomes narrower. Therefore, the use of narrow band light enables graphene with narrow layer range to be rapidly distinguished. For example, we can easily sort out graphene with less than 10 layers by using a light of 430–570 nm. These results suggest that the selection of an appropriate wavelength range of light source is very helpful for the rapid thickness evaluation and accurate layer identification.

We also calculated the TCD as a function of the number of graphene layers and Al_2O_3 film thickness for a preferential light source of 430–570 nm and a random light source of 500–600 nm, shown in Figure 4a,b. It can be found that the optimum substrate is 70 and 210 nm thick Al_2O_3 film for the light source of 430–570 nm but is 65 and 197 nm thick Al_2O_3 film for 500–600 nm light source. These are also different from the 72 and 185 nm thick Al_2O_3 film for halogen light of 380–720 nm in Figure 2a. These results reveal that, if the light source is different, the optimum substrates are also different. Therefore, the TCD method also provides an easy way to forecast the optimum substrate for a specific wavelength of light.

It is worth noting that all the above calculated results are based on the orthogonal incidence. Actually, the incidence and reflection of light on samples are not orthogonal in a real-world optical system with a finite numerical aperture (NA),⁶ and they are in fact conical (Figure 5a) as quantified by the NA of the objective lens. The maximal incident angle, θ_{max} , can be determined by the equation $\text{NA} = n \sin \theta_{\text{max}}$ where n is the refraction rate of the medium between objective lens and specimen. Here, we adopt $n = 1$ because the medium

is air in our experiments. In this case, different interference patterns are obtained depending on the incident angle, and they are summed up incoherently in the OM due to the typical poor coherence of the exploited light sources. Consequently, this will generally lead to a complex light averaging. Assuming that the intensity profiles of reflected light along the angle of incidence follow a Gaussian distribution,¹⁴ the total reflectivity of light can be obtained by integrating each component throughout the angle of incidence ($-\theta_{\text{max}} - \theta_{\text{max}}$):

$$R = \int_{-\theta_{\text{max}}}^{\theta_{\text{max}}} R(\theta) dN(\theta, 0, \theta_{\text{max}}/3) \quad (6)$$

where $R(\theta)$ represents the reflectivity for the incident angle of θ , and $dN(\theta, 0, \theta_{\text{max}}/3)$ represents the Gaussian distribution with the mean of 0 and standard deviation of $\theta_{\text{max}}/3$, at an angle interval of $d\theta$.

Take the $\text{Al}_2\text{O}_3/\text{Si}$ substrate as an example; we calculated the TCD values of graphene layers on the Si substrate with different thicknesses of Al_2O_3 films for four commonly used objective lens of 10 \times , 20 \times , 50 \times , and 100 \times (corresponding to the NA values of 0.3, 0.45, 0.8, and 0.9, respectively), and the results are shown in Figure 5b–e, respectively. It can be found that the TCD values change slightly, and all the preferential thickness of Al_2O_3 for these four objectives is 72 nm. Figure 5f shows the detailed TCD values of graphene layers on the preferential substrate of 72 nm thick $\text{Al}_2\text{O}_3/\text{Si}$. It is needed to point out that the TCD inflexions are almost the same for these four cases. The smaller the NA of the objective, the larger the TCD values for the graphene less than 15 layers, while the smaller the NA of the objective, the smaller the TCD values for the graphene with layers in the range of 16–35. The final TCD values change very slightly for orthogonal incidence and different objective lens due to the Gaussian distribution of reflected light for different angles. The maximum TCD variation is smaller than 1.0 for the two ultimate cases

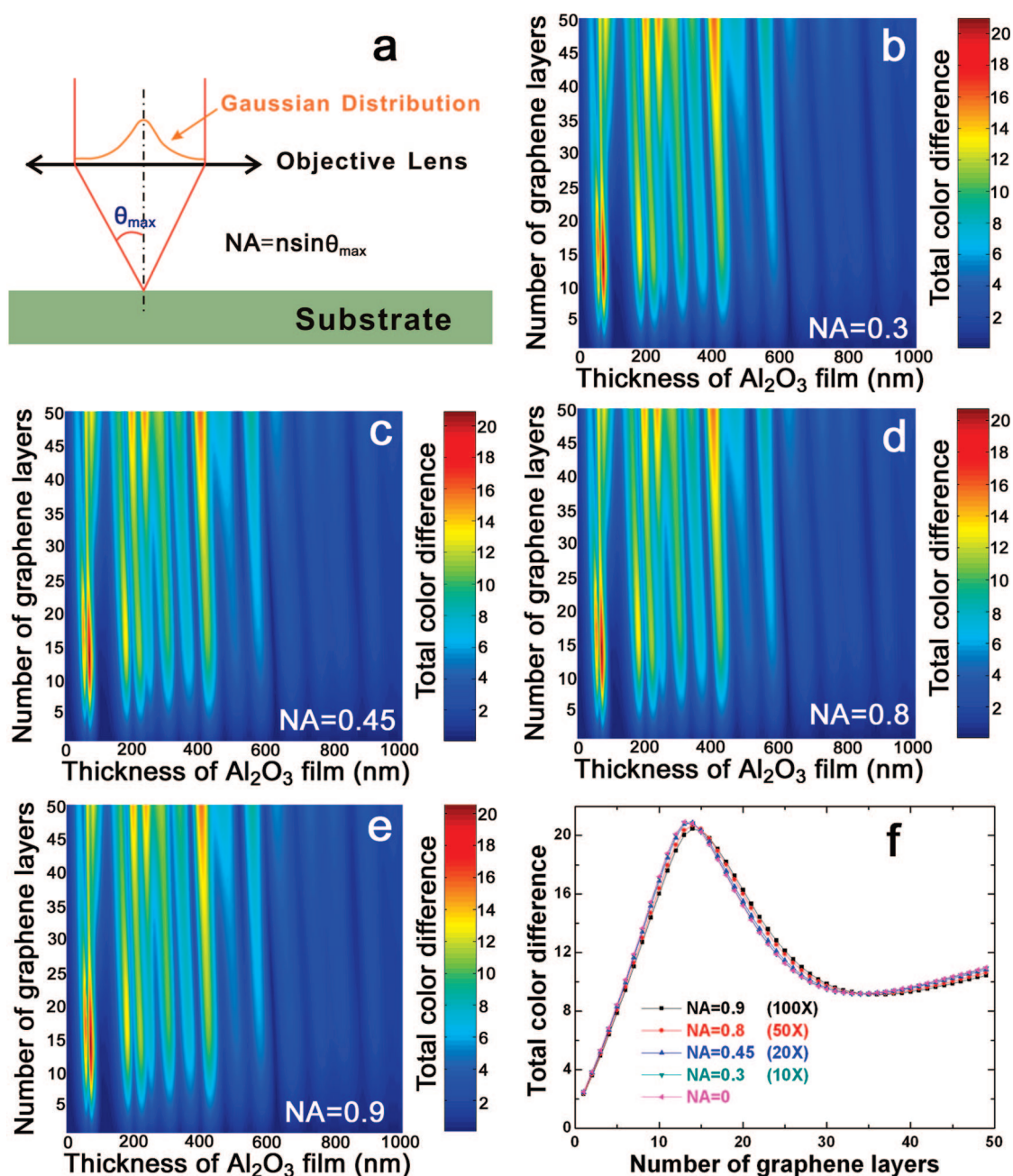


Figure 5. (a) Optical path through the objective lens. Calculated TCD values of graphene as a function of the number of graphene layers and the thickness of Al_2O_3 film for objective lenses of (b) $\text{NA} = 0.3$, (c) $\text{NA} = 0.45$, (d) $\text{NA} = 0.8$, and (e) $\text{NA} = 0.9$. (f) Detailed TCD variation of graphene on the preferential substrate of Si capped with a 72 nm thick Al_2O_3 film for different objective lens.

of $\text{NA} = 0.9$ and orthogonal incidence. The calculated TCD values with the assumption of orthogonal incidence are still valid for the identification of graphene layers experimentally obtained by the OM with a different objective lens. Of course, the same imaging conditions as those adopted in OM should be used in the TCD calculation for the better identification of graphene layers.

By improving the image contrast of graphene guided by our calculated results, we can easily ascertain the number of graphene layers simply by compar-

ing the calculated (orthogonal incidence) and experimentally obtained TCD values. To demonstrate it, graphene samples were prepared by the micromechanical cleavage method and transferred to Si substrates with a 72 nm thick Al_2O_3 film.⁵ Figure 6a shows the optical image of graphene under halogen light, and it is clear that the graphene layers at different positions (marked 1, 2, 3, 4, 5, 6, and 7) exhibit different colors with well-distinguished borders. By comparing the apparent color of graphene in Figure 6a with the calculated color bar shown in Figure 6b, the graphene lo-

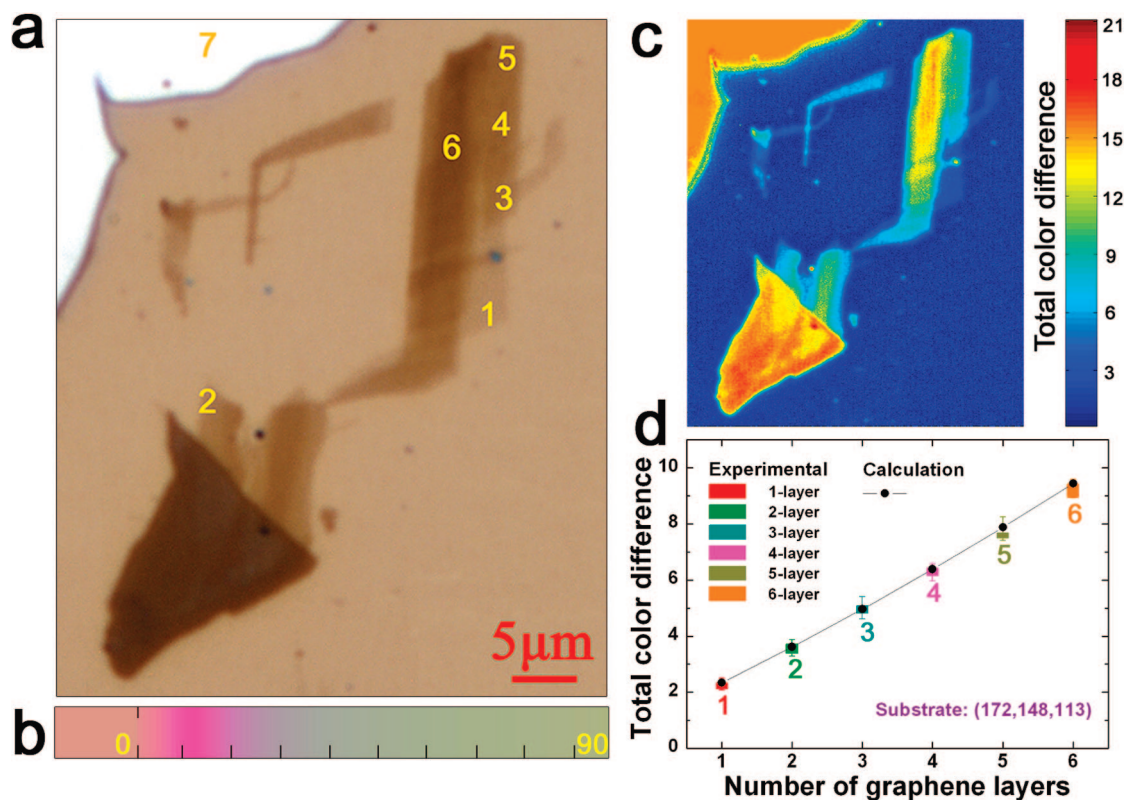


Figure 6. (a) Optical image of graphene on the Si substrate with a 72 nm thick Al_2O_3 film. Graphene with different apparent colors were marked with 1, 2, 3, 4, 5, 6, and 7. (b) Calculated color bar of graphene on the Si substrate with a 72 nm thick Al_2O_3 film, and the number corresponds to the number of graphene layers. (c) Experimentally derived TCD values of graphene in the entire area of (a) from their corresponding RGB data. (d) Comparison between theoretically predicted TCD values for different layers and experimentally obtained TCD values from the marked positions 1–6 in (a), the numbers in parentheses are the RGB value of the substrate. In order to minimize errors, we collected the RGB data from several spots close to the same position and adopted the average of the corresponding TCD values as the experimentally derived TCD for comparison.

cated at positions 1–6 can be easily assigned to be less than 12 layers. To further identify the layers of graphene with different apparent colors, we collected some corresponding RGB data from Figure 6a and transformed them into TCD values by eq5. Figure 6c presents the processed three-dimensional TCD values of graphene in the entire area of Figure 6a, which clearly shows a good agreement with the real image. The TCD values of graphene at positions 1–6 derived from RGB data were compared with the calculated TCD values (obtained from Figure 2d) of graphene with different layers (Figure 6d). It is necessary to point out that, in order to minimize errors, we collected the RGB data from several spots close to the same position and adopted the average of the corresponding TCD values as the experimentally derived TCD for comparison. The good agreement between experimentally derived and calculated values indicates that graphene at positions 1–6 is 1, 2, 3, 4, 5, and 6 layers, respectively. We suggest that the tiny difference between experimentally derived and calculated values is attributed to the different incidence conditions, as discussed above.

For testing the above layer assignments by TCD method, we used Raman spectroscopy to characterize the layers of graphene at the marked positions in Fig-

ure 6a, and the corresponding Raman spectra are shown in Figure 7. According to the position and profile of the 2D band and the intensity of the G band,^{8–11} we conclude that the graphene in marked positions

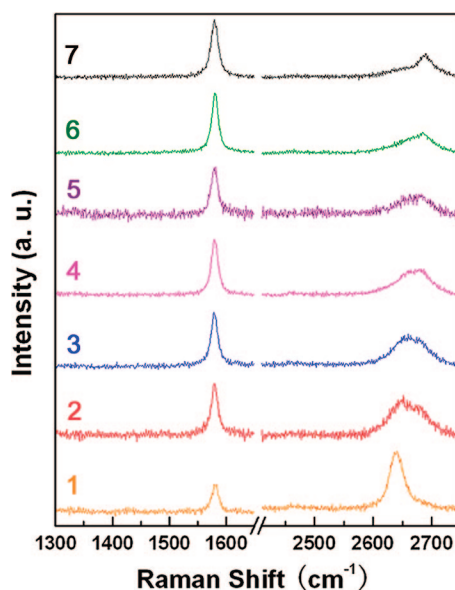


Figure 7. Raman spectra of graphene marked with 1, 2, 3, 4, 5, 6, and 7 in Figure 6a, respectively, from bottom to top.

1–6 are 1, 2, 3, 4, 5, and ~6 layers. This is in good agreement with that obtained by the TCD method, indicating that the TCD method is a feasible and effective way to identify the layers of graphene.

It should be noted that Al_2O_3 is a commonly used gate material for field effect transistors (FETs); therefore, our results indicate that the direct fabrication and property measurements of graphene-based electronics in OM is possible, avoiding the damage of graphene by electron irradiation in SEM. Additionally, this method can be easily extended to predict the visibility of graphene located on the top of other potential dielectric films¹⁸ suitable for the fabrication of electronics, and even substrates of different wafers (instead of Si, such as Ag, Au, Ni, or Cu)¹⁹ covered by dielectric multiple films. Furthermore, this method can also be used as a universal one for the characterization of graphene materials prepared by other methods. However, we should point out that some parameters in the calculations, such as the refractive index and the thickness of graphene, should be changed for graphene materials prepared by other methods. For example, graphene prepared by chemical exfoliation has different refractive index and different thickness from that prepared by mechanical cleavage since there are some functional groups on their edges and surfaces.¹⁴

EXPERIMENTAL SECTION

The graphene samples were prepared by a micromechanical cleavage method and transferred to the substrates.⁵ Si substrates with different thickness of dielectric film were made by a film coating system (Gatan Model 682 precision etching coating system). The colorful images of graphene samples and substrate were taken using a Carl Zeiss Axiovert 200 MAT Inverted OM with a 12 V 100WHA1-L lamp (Philips 7724 and operated at 7 V). The Raman spectra were measured and collected using a 632.8 nm laser under ambient conditions with JY HR800, and a laser power below 0.1 mW was used in our Raman measurements to avoid laser-induced heating.

Acknowledgment. The authors thank C. Y. Zhang and J. Tan for the support on OM measurements, and C. Liu and S. S. Li for the kind help on film coating. This work was supported by the Ministry of Science and Technology of China (No. 2006CB932701), National Science Foundation of China (No. 90606008 and No. 50702063), Chinese Academy of Sciences (No. KJCX2-YW-M01), and the Knowledge Innovation Program of CAS.

Supporting Information Available: Optical image, calculated color bar and TCD of graphene on Si with a 285 nm thick SiO_2 film, and comparisons of the wavelength spectra of halogen and standard D65 light and the calculated color and TCD. This material is available free of charge via the Internet at <http://pubs.acs.org>.

REFERENCES AND NOTES

- Geim, A. K.; Novoselov, K. S. The Rise of Graphene. *Nat. Mater.* **2007**, *6*, 183–191.
- Zhang, Y. B.; Tan, Y. W.; Stormer, H. L.; Kim, P. Experimental Observation of the Quantum Hall Effect and Berry's Phase in Graphene. *Nature* **2005**, *438*, 201–204.
- Novoselov, K. S.; Jiang, Z.; Zhang, Y.; Morozov, S. V.; Stormer, H. L.; Zeitler, U.; Maan, J. C.; Boebinger, G. S.; Kim,

CONCLUSIONS

We have proposed a TCD method for the rapid and accurate identification of graphene and to effectively guide the selection of substrate and light source for characterization and determination of graphene layers by optical images based on a combination of reflection spectrum and CIE color space. The preferential thickness and type of dielectric films on a Si substrate for the observation of graphene in white light were well elucidated. We find that a 72 nm thick Al_2O_3 film on the Si substrate is much better than the commonly used SiO_2/Si and $\text{Si}_3\text{N}_4/\text{Si}$ substrates, and the image contrast and TCD can be enhanced if the wavelength range of light source is narrowed appropriately. Our results also show that a monochromatic light source is not the best choice for the identification of graphene. The TCD values for the same graphene change slightly with different objective lenses in a real-world optical system. Moreover, it is demonstrated that the number of graphene layers can be rapidly and accurately identified by comparing the experimentally derived TCD values from the image and the calculated TCD values. These findings will be very helpful for the selective preparation of graphene with predetermined layers and open up the possibility for the nondestructive characterization and physical property measurements of graphene-based devices under OM.

- .; Geim, A. K. Room-Temperature Quantum Hall Effect in Graphene. *Science* **2007**, *315*, 1379.
- Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.; Zhang, Y.; Dubonos, S. V.; Grigorieva, I. V.; Firsov, A. A. Electric Field Effect in Atomically Thin Carbon Films. *Science* **2004**, *306*, 666–669.
 - Novoselov, K. S.; Jiang, D.; Schedin, F.; Booth, T. J.; Khotkevich, V. V.; Morozov, S. V.; Geim, A. K. Two-Dimensional Atomic Crystals. *Proc. Natl. Acad. Sci. U.S.A.* **2005**, *102*, 10451–10453.
 - Rodaro, S.; Pingue, P.; Piazza, V.; Pellegrini, V.; Beltram, F. The Optical Visibility of Graphene: Interference Colors of Ultrathin Graphite on SiO_2 . *Nano Lett.* **2007**, *7*, 2707–2710.
 - Meyer, J. C.; Geim, A. K.; Katsnelson, M. I.; Novoselov, K. S.; Booth, T. J.; Roth, S. The Structure of Suspended Graphene Sheets. *Nature* **2007**, *446*, 60–63.
 - Gupta, A.; Chen, G.; Joshi, P.; Tadigadapa, S.; Eklund, P. C. Raman Scattering from High-Frequency Phonons in Supported n-Graphene Layer Films. *Nano Lett.* **2006**, *6*, 2667–2673.
 - Ferrari, A. C.; Meyer, J. C.; Scardaci, V.; Casiraghi, C.; Lazzeri, M.; Mauri, F.; Piscanec, S.; Jiang, D.; Novoselov, K. S.; Roth, S. Raman Spectrum of Graphene and Graphene Layers. *Phys. Rev. Lett.* **2006**, *97*, 187401.
 - Graf, D.; Molitor, F.; Ensslin, K.; Stampfer, C.; Jungen, A.; Hierold, C.; Wirtz, L. Spatially Resolved Raman Spectroscopy of Single- and Few-Layer Graphene. *Nano Lett.* **2007**, *7*, 238–242.
 - Graf, D.; Molitor, F.; Ensslin, K.; Stampfer, C.; Jungen, A.; Hierold, C.; Wirtz, L. Raman Imaging of Graphene. *Solid State Commun.* **2007**, *143*, 44–46.
 - Blake, P.; Hill, E. W.; Neto, A. H. C.; Novoselov, K. S.; Jiang, D.; Yang, R.; Booth, T. J.; Geim, A. K. Making Graphene Visible. *Appl. Phys. Lett.* **2007**, *91*, 063124.

13. Ni, Z. H.; Wang, H. M.; Kasim, J.; Fan, H. M.; Yu, T.; Wu, Y. H.; Feng, Y. P.; Shen, Z. X. Graphene Thickness Determination Using Reflection and Contrast Spectroscopy. *Nano Lett.* **2007**, *7*, 2758–2763.
14. Jung, I.; Pelton, M.; Piner, R.; Dikin, D. A.; Stankovich, S.; Watcharotone, S.; Hausner, M.; Ruoff, R. S. Simple Approach for High-Contrast Optical Imaging and Characterization of Graphene-Based Sheets. *Nano Lett.* **2007**, *7*, 3569–3575.
15. Janos, S. *Colorimetry: Understanding the CIE System*; John Wiley & Sons: New Jersey, 2007; pp 25–88.
16. Heavens, O. S. *Optical Properties of Thin Solid Films*; Dover Publications: New York, 1965.
17. Henrie, J.; Kellis, S.; Schultz, S. M.; Hawkins, A. Electronic Color Charts for Dielectric Films on Silicon. *Opt. Express* **2004**, *12*, 1464–1469.
18. Nalwa, H. S. *Handbook of Low and High Dielectric Constant Materials and Their Applications*; Academic Press: New York, 1999.
19. Palik, E. D. *Handbook of Optical Constants of Solids II*; Academic Press: Boston, MA, 1991.