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Determination of hBN Thickness by Optical Contrast

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Abstract

Van der Waals materials are crystals that can be peeled into atomically thin layers. When a number of different layer types stack upon one another, different physical properties appear. A commonly studied van der Waals material is hexagonal boron nitride (hBN). Hexagonal boron nitride is highly stable and has a structure similar to that of graphite. Such atomic arrangement allows hBN to be extremely hard, have novel electrical properties, and excellent thermal conductivity. However, whenever hBN layers are stacked upon one another, the physical and optical properties of the crystal differ with thickness, and so with the number of hBN layers. By using Fresnel's equations, a plot function was coded in MATLAB that compared a range of wavelengths with a range of hBN thicknesses. The ranges were plotted against the values of the light contrast from the samples. The plot was tested experimentally by shining 6 different colored LEDs with different wavelengths, at normal incidence, onto a sample made up of an unknown number of hBN layers on a Silicon (Si) chip with a 300 nm thermally grown oxide layer. A microscope was used to capture images of the different samples. The images were transferred into a python program that measured the contrast of the grayscale images and returned the thickness of the samples. The results of the code were compared to the thickness measurements of the sample received by an atomic force microscope (AFM). Flake 1 was measured using the AFM for a thickness of 5 nm and the equations using the image contrasts calculated a thickness of 3 nm. Flake 2 was measured using the AFM for a thickness of 14 nm and the equations using the image contrasts calculated a thickness of 18 nm.

1. Introduction

Van der Waals (vdW) materials are crystals consisting of weakly coupled 2D planes which can be exfoliated into layers, in some cases down to atomic thicknesses. The isolation of graphene [1], a single layer of carbon atoms arranged in a honeycomb lattice, sparked the study of these materials. Today they are prized for their extreme thinness and novel electronic and optical properties.

Hexagonal boron nitride (hBN) consists of a honeycomb lattice of alternating boron and nitrogen atoms and can be exfoliated down to atomically thin layers [2]. The material is interesting in its own right as a wide band semiconductor [3]–[5], and has found a critical niche in the vdW field as an insulating layer in heterostructures made of several materials [6]–[9]. Consequently, hBN may play an important role as an insulator, atomically-flat substrate, and/or protective layer in future vdW devices such as photodetectors and transistors [10]–[12].

It is notable that atomically thin materials can be seen at all with only the aid of a light microscope. The study of the optical contrast of vdW materials began with graphene in 2007 shortly after its isolation [13]. By using the Fresnel equations and silicon substrates with precisely known oxide layers, we can determine the thickness of deposited flakes. This in term provides the number of atomic layers in the flake, information critical to the properties and applications of vdW materials. This method has been widely used to study graphene [13], [14], transition metal dichalcogenides [15]–[17], and previous studies of hBN itself [18]–[21] and is the basis for emerging approaches to automated flake searches and layer number determination [17], [22], [23].

Here, we present a study of the contrast of hBN on Si/SiO_2 substrates, establishing flake thicknesses via a Fresnel model and comparing them to thickness results from atomic force microscopy. Our methodology is notable for its ease of implementation, eschewing narrow band optical filters [18], [20], complex optics [19], and RGB decomposition of images [17], [21] for inexpensive LEDs with known peak wavelengths. This is a step forward in the democratization of the study of vdW materials.

2. Theoretical Modeling

A Fresnel model was used to determine the contrast between a flake of hBN on a Si/SiO_2 substrate and the bare substrate itself. The Fresnel-law-based model [24] uses an equation of reflected light intensity, off of multiple layers, to calculate contrast of reflected light between regions with a crystal and without. The equation was noted to be easily derived to fit multiple layers of incidence stacked on top of one another by changing electric field (E) equations with matrix forms. We begin from matching the E-field above and below each interface:

$$E_n = E_{in} + E_{rn} = E_{tn} + E'_{r(n+1)},$$

where n represents the layer count, *i* indicates E-field from an incident ray, r from a reflected ray, and t from a transmitted ray. The prime indicates a phase shift across the thickness of the layer. The light's magnetic fields (H) are described in the terms of electric fields in the form:

$$H_n = \sqrt{\frac{\epsilon_0}{\mu_0}}(E_{in} - E_{rn})n_{n-1}cos\theta_{in} \ ,$$

where n_n is the index of refraction of the nth material. However, due to the angle of light in this experiment being perpendicular to the surface of the sample (0°) we remove the cosine from the term. In addition, while the light passes the layers of incidence, a phase shift on its electric field occurs that is represented as:

$$E_{in} = E_{r(n-1)}e^{-i\theta_{n-1}},$$

$$E_{rn} = E'_{rn}e^{+i\theta_{n-1}}.$$

Limiting ourselves to three layers for demonstration, by using Euler's identity and combining the equations for three (n = 1,2,3) layers, we received the two following linear relations:

$$E_1 = E_2\cos(\theta_1) + \ H_2(i\sin(\theta_1))/Y_1 \qquad E_2 = E_3\cos(\theta_1) + \ H_3(i\sin(\theta_2)/Y_2$$

$$H_1 = E_2 Y_1 i\sin(\theta_1) + H_2\cos(\theta_1) \qquad H_2 = E_3 Y_2 i\sin(\theta_2) + H_3\cos(\theta_1)$$

where $Y_n = \sqrt{\frac{\epsilon_0}{\mu_0}}n_n$.

The matrix notation for the two linear relations above can be written as

$$\begin{bmatrix} E_1 \\ H_1 \end{bmatrix} = \begin{bmatrix} \cos\ \theta_1 & (i\sin(\theta_1))/Y_1 \\ (i\sin(\theta_1))/Y_1 & \cos\ \theta_1 \end{bmatrix} \begin{bmatrix} E_2 \\ H_2 \end{bmatrix} \ and \ \begin{bmatrix} E_2 \\ H_2 \end{bmatrix} = \begin{bmatrix} \cos\ \theta_2 & (i\sin(\theta_2))/Y_2 \\ (i\sin(\theta_2))/Y_2 & \cos\ \theta_2 \end{bmatrix} \begin{bmatrix} E_3 \\ H_3 \end{bmatrix}$$

This relation can be stacked with an arbitrary number of layers, and it results into the reflection coefficient (r) by multiplying the matrixes together. The form follows the pattern

$$\begin{bmatrix} E_1 \\ H_1 \end{bmatrix} = \begin{bmatrix} \cos\ \theta_1 & (i\sin(\theta_1))/Y_1 \\ (i\sin(\theta_1))/Y_1 & \cos\ \theta_1 \end{bmatrix} \begin{bmatrix} \cos\ \theta_2 & (i\sin(\theta_2))/Y_2 \\ (i\sin(\theta_2))/Y_2 & \cos\ \theta_2 \end{bmatrix} \begin{bmatrix} E_3 \\ H_3 \end{bmatrix}$$

Now, the electric and magnetic field equations that are emitted from the light source can be replaced with their reflected and transmitted values to become

$$\begin{bmatrix} E_{i1} + E_{r1} \\ (E_{i1} - E_{r1})/Y_0 \end{bmatrix} = M \begin{bmatrix} E_{t3} \\ E_{t3} Y_3 \end{bmatrix}$$

The matrix equations describe the two relations:

$$E_{i1} + E_{r1} = m_{11} E_{t3} + m_{12} E_{t3} Y_3$$

$$(E_{i1} - E_{r1})/Y_0 = m_{21} E_{t3} + m_{22} E_{t3} Y_3$$

where m_{ab} is an element of M. Solving for the reflection coefficient relationship, the result becomes:

$$r = \frac{Y_0 m_{11} + Y_0 Y_3 m_{12} - m_{21} - Y_3 m_{22}}{Y_0 m_{11} + Y_0 Y_3 m_{12} + m_{21} + Y_3 m_{22}}$$

From this, we can determine the reflectance off of the layers, $R=r^2$, which is proportional to the incoming intensity of light. If the same intensity of light is used to illuminate the region with a flake of interest as is used to illuminate an adjacent region without the flake, then the contrast can be written as:

$$C = \frac{r^2_{\text{without hBN}} - r^2_{\text{with hBN}}}{r^2_{\text{without hBN}}}$$

The contrast C equation was implemented using MATLAB with a semi-infinite slab of Si (~1mm), a thin layer of SiO_2 (300nm), and an unknown hBN thickness. Using the reflection coefficient equation, a density contrast plot, Figure 1, was programmed as a function of hBN thickness (nm) and wavelength of light (nm).

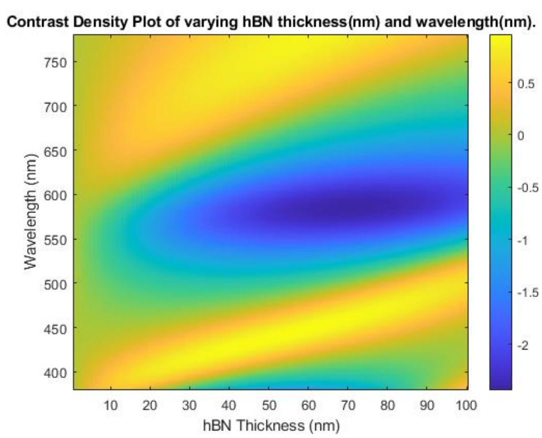

Figure 1. Contrast density plot of the hBN on an SiO_2/Si wafer representing contrast with a color bar. The yellow-orange colors represent high contrast with a bright wafer whereas blue colors represent high contrast with a bright hBN flake. Green-yellow colors are low contrast.

3. Experimental Methods

The extracted hBN crystals were mechanically exfoliated onto the Si/SiO_2 chips in the form displayed by Figure 2a [2]. The crystals were visible through a microscope with a number of colors due to the existence of different thicknesses of hBN crystals. Two thin light green flakes were selected from the Si/SiO_2 chip for the experiment. Six images were taken for both flakes using six different known LED wavelengths. The peak LED wavelengths were measured using a spectrometer with good agreement to nominal values: 457nm, 473nm, 526nm, 592nm, 613nm, and 633nm.

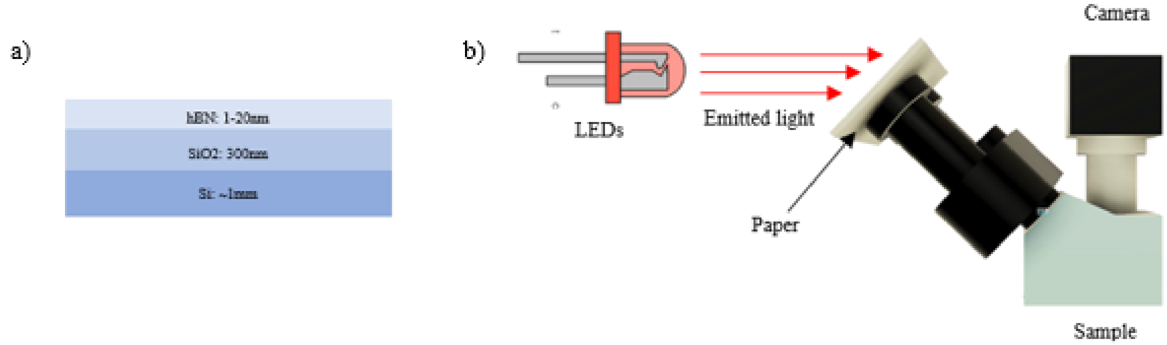

Figure 2. a) A side-view of the wafer sample layers with the measurements of their thicknesses. b) The microscope setup for the images consisted of six LEDs emitting light through a white piece of paper on top of the microscope lens that diffused the light.

The light by the LEDs was diffused by a white piece of paper onto the lens of a microscope whose structure bent the light perpendicularly onto the stage. This enabled the implementation of the Fresnel model derived in section 2. In addition, the experiment was hosted inside a dark room where light could not interfere with the LED wavelengths to reduce any contrast inflicted by ambient light. Images were taken for both flakes using a camera equipped with the microscope as in Figure 2b.

5. Results

The experiment created twelve images total, six images of flake one and six images of flake two. Each picture was taken under different LED wavelengths shown in Figure 3. The images were then gray scaled and processed with a Python program that calculated the contrast values between the average Si/SiO_2 chip pixels and the average pixels of the hBN flakes.

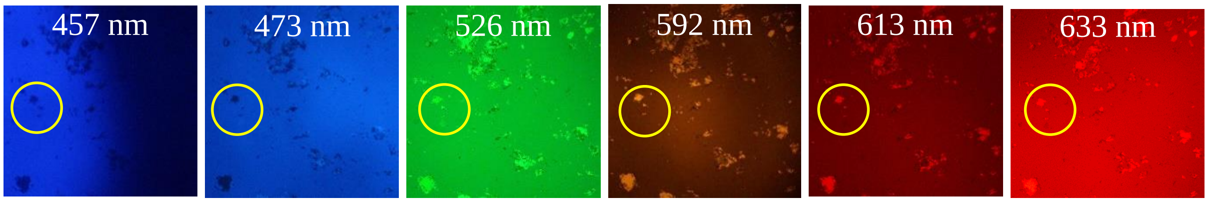

Figure 3. Flake 1 is located at the yellow highlighted locations above. The wavelengths emitted by the LEDs are displayed on each image. The illumination varies across the Si/SO2 chip due to the methodology; however, the illumination is high and approximately constant at the location of the flake. Similar images were taken for flake 2.

A data set of 600 contrasts was then extracted from the density plot from Figure 1 for each of the six wavelengths with a varying hBN thickness of 1 nm to 100 nm. A least-squares residual plot was calculated using Equation 1 to find the closest hBN thickness that matched the data from the images. The plots can be seen in Figure 4a and Figure 4b where the lowest residual for flake 1 was closest to a hBN thickness of 3nm and the lowest residual for flake 2 was closest to a hBN thickness of 18nm.

$$\sum_{n=1}^{6}(D(\lambda_i) - C(d\,,\lambda_i))^2$$

Equation 1. λ_i represents LED wavelengths, d represents hBN layer thickness, D(λ_i) represents the flake contrast at the six wavelengths, and C(d, λ_i) represents the contrast between 1-100 nm thickness for all six wavelengths.

The hBN thicknesses were used as estimations of the actual flake samples. The six contrast values from the images in Figure 3 were plotted against the density plot values of these thickness estimations; flake 1 contrasts are compared in Figure 4c, and flake 2 contrasts are compared in Figure 4d. Additionally, the thickness values for both flakes were measured with an AFM in Figure 5. Flake 1 had an average thickness of 5nm and flake 2 had an average thickness of 14nm.

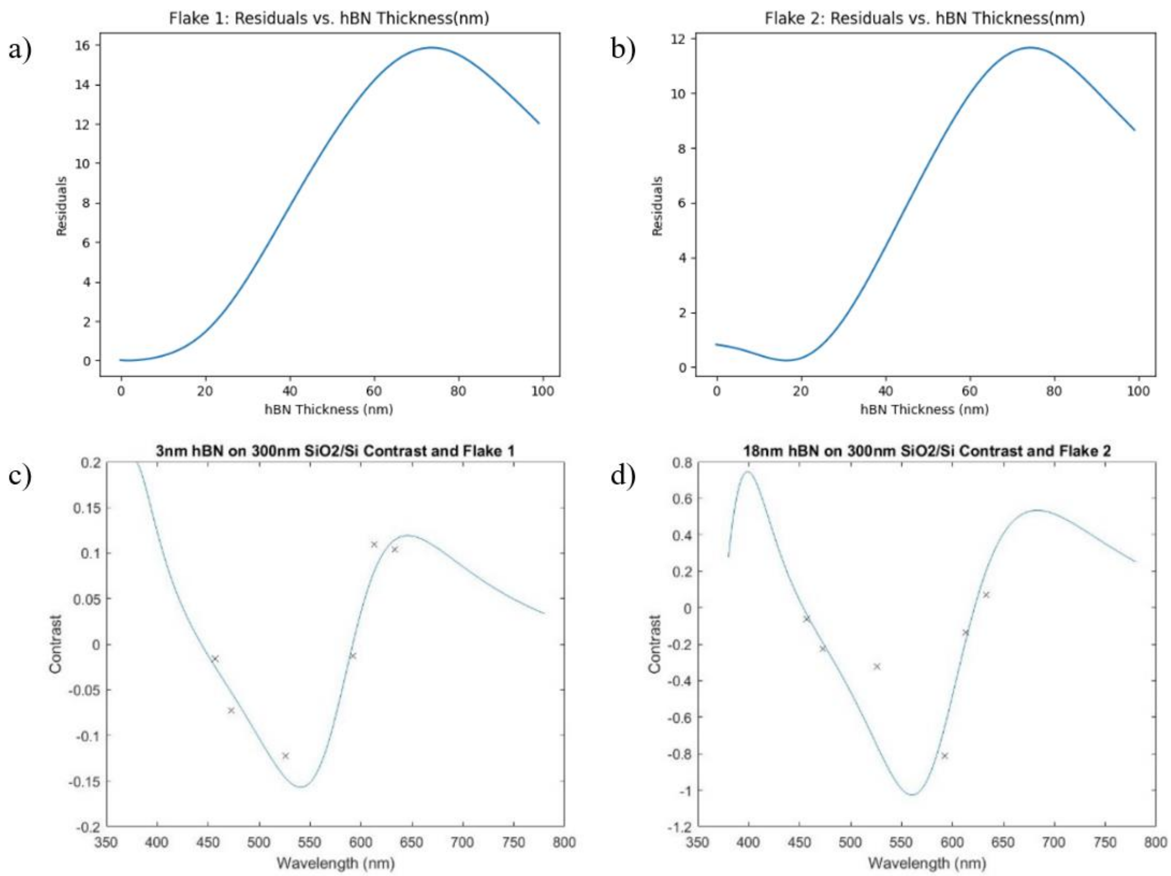

Figure 4. a) Flake 1 residual plot calculated from the python program where the lowest residual value is located at 3nm hBN thickness as 0.00218. b) Flake 2 residual plot has the lowest residual value located at 18nm hBN thickness 0.2356 c) A comparison between the contrast values calculated through the microscope images for flake 1 and the contrast density plot values in Figure 1 when hBN has a thickness of 3 nm. d) A comparison between the contrast values calculated for flake 2 and the contrast density plot values in Figure 1 when hBN has a thickness of 18 nm.

a) b)

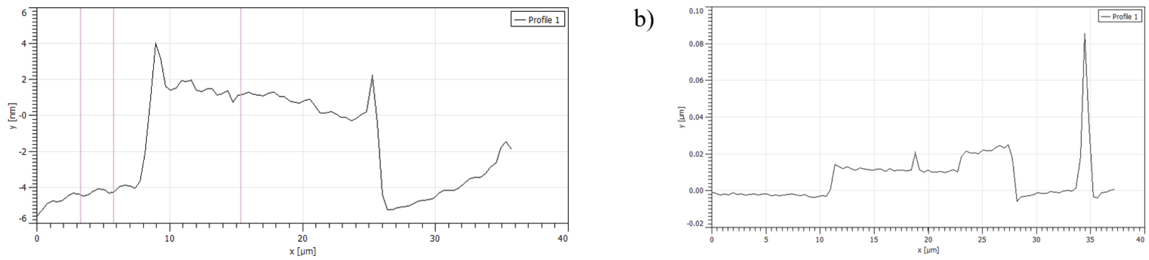

Figure 5. a) An AFM data set for the thickness of flake 1 with an average of 5nm. b) An AFM data set for the thickness of flake 2 with an average of 14nm.

5. Discussion

The investigation demonstrates a new methodology of measuring the thickness of hexagonal boron nitride. The 2 nm and 4 nm differences between the model and the AFM results are small in magnitude but significant. They are reflective of limitations in the methodology, especially the width of the LED emission peaks, but they demonstrate that this experiment can be completed reasonably with readily available materials and easily understood calculations.

The peak values from the LED spectra were used for the python program. By accounting for the entire spectrum of the LED lights [17], it may be possible to decrease the difference between the AFM results and the model in Figure 1. Likewise, the LED setup could be improved to emit a more uniform illumination of the substrate.

Further implementation with additional materials, as well as improvements on the experimental design, may lead to easier measurements of two-dimensional material thicknesses and assist in developing a methodology of calculating the number of singular layers of vdW materials on Si/SiO_2 substrates.

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7. Citations

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