

# Optical Imaging and Characterization of Graphene and Other 2D Materials Using Quantitative Phase Microscopy

Samira Khadir,<sup>\*,†</sup> Pierre Bon,<sup>‡</sup> Dominique Vignaud,<sup>§</sup> Elizabeth Galopin,<sup>§</sup> Niall McEvoy,<sup>⊥,||,¶</sup> David McCloskey, <sup>[], ¶, #</sup> Serge Monneret,<sup>†</sup> and Guillaume Baffou<sup>\*,†</sup>

<sup>†</sup>Institut Fresnel, CNRS, Aix Marseille Univ, Centrale Marseille, Marseille, France

<sup>‡</sup>Laboratoire Photonique Numérique et Nanoscience (LP2N), CNRS UMR5298, Bordeaux University, Institut d'Optique Graduate School, Talence 33405, France

<sup>§</sup>IEMN, University of Lille, Avenue Poincaré CS 60069, Villeneuve d'Ascq Cedex 59652, France

<sup>1</sup>School of Chemistry, Trinity College Dublin, Dublin 2, Ireland

<sup>II</sup>Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN), Trinity College Dublin, Dublin 2, Ireland

<sup>¶</sup>Advanced Materials and BioEngineering Research (AMBER), Trinity College Dublin, Dublin 2, Ireland

<sup>#</sup>School of Physics, Trinity College Dublin, Dublin 2, Ireland

Supporting Information

ABSTRACT: This article introduces an optical microscopy technique for the characterization of two-dimensional (2D) materials. The technique is based on the use of quadriwave lateral shearing interferometry (QLSI), a quantitative phase imaging technique that allows the imaging of both the intensity and the phase of an incoming light beam. The article shows how QLSI can be used to (i) image 2D materials with high contrast on transparent substrates, (ii) detect the presence of residues coming from the fabrication process, and (iii) map the 2D complex optical conductivity and complex refractive index by processing the intensity and phase images of a light beam crossing the 2D material of interest. To illustrate the versatility of this approach for 2D material imaging and characterization, measurements have been performed on graphene and MoS<sub>2</sub>.



**KEYWORDS:** wavefront sensing, phase imaging, 2D materials, graphene, MoS<sub>2</sub>, optical conductivity, refractive index

raphene is a single-atom-thick, two-dimensional (2D) material composed of carbon atoms arranged in a honeycomb lattice structure. Due to its fascinating properties including massless Dirac fermions,<sup>1</sup> anomalous quantum Hall effect,<sup>1,2</sup> and high intrinsic strength,<sup>3</sup> graphene is already at the basis of various promising applications, such as spintronics,<sup>4</sup> transparent electrodes for solar cells and light display technologies,<sup>5</sup> supercapacitors,<sup>6</sup> among others.<sup>7-9</sup> Since the first isolation of graphene in 2004,10° a large variety of 2D materials such as semiconducting transition metal dichalcogenides  $(MoS_2, MoSe_2, WSe_2, ...)$  have been synthesized. All these 2D materials are considered as promising candidates for generating novel technological applications in various domains.<sup>11,1</sup>

The geometrical and optical characterizations of 2D materials are not straightforward, as they are quite transparent and their thickness does not exceed the nanometer scale. Several methods have been used for determining the number of graphene layers. For instance, atomic force microscopy (AFM) is widely used to probe and image the thickness of 2D materials.<sup>10</sup> However, this approach is delicate and timeconsuming. Raman spectroscopy is another widely used

technique to characterize graphene.<sup>13</sup> Although this technique allows the identification of single-layer (SL) graphene from the far field, it is not robust to identify the number of layers if greater than 2. In addition, this method can be invasive, as it requires the use of a focused laser beam that can sometimes reach 1 mW.<sup>14</sup> Such a large power results in a large irradiance (around 10<sup>7</sup> mW·cm<sup>-2</sup>) that can induce the formation of defects modifying the optical properties of the material.<sup>14,15</sup> Recently, other optical methods have been proposed to identify the number of layers of 2D materials. Among them, one can simply use an optical microscope and measure the image contrast in reflection or in transmission.<sup>16,17</sup> But this method is not reliable, as the contrast differs from one experiment to another and the contrast is very weak in the case of SL graphene on a transparent substrate.<sup>18</sup> SiO<sub>2</sub>-coated silicon wafers are usually mandatory to make SL graphene visible.<sup>19-21</sup> The surface plasmon polariton (SPP) property of noble metals

Special Issue: 2D Materials for Nanophotonics

Received: July 28, 2017 Published: September 27, 2017





Figure 1. (a) Scheme of a QLSI microscope setup. A Köhler illumination is used to illuminate the sample with a light beam controlled in size and numerical aperture. A band-pass filter is used to select the spectral range of interest. The sample is composed of microscale objects (2D materials in the context of this article). The wavefront analyzer is constituted by a modified Hartmann grating (MHG) and a CCD camera. (b) Scheme of the principle of the QLSI technique. A refractive object characterized by its refractive index  $\underline{n}$  deposited on a substrate having a refractive index  $n_2$  and surrounded by a medium of a refractive index  $n_1$  causes a distortion of an incoming planar wavefront.

has also been proposed as a means to determine the number of graphene layers.<sup>22</sup> This method requires the deposition of a thin layer of noble metal under the graphene sheets to generate the SPP in the visible range. Additional technological steps are needed, and the presence of the metal layer makes the sample opaque and may modify the properties of the graphene deposited on top.

In addition to their limitations cited above, these methods only allow the identification of the number of layers of the 2D material without giving information about the physical properties of the studied material (complex refractive index, dielectric function, etc.). Other techniques are generally required to measure the optical response of 2D materials, e.g., frequency-dependent transmittance and reflectance measurements,<sup>23,24</sup> picometrology,<sup>25</sup> and spectroscopic ellipsometry.<sup>26</sup> Spectroscopic ellipsometry can also operate in an imaging mode.<sup>27,28</sup> This approach is powerful to image, classify, and determine the optical properties of 2D materials on the microscale. However, this technique uses an unconventional, specific tilted illumination that cannot be easily implemented in standard microscopes. In addition, such an illumination limits the lateral resolution to about 1  $\mu$ m. In 2010, Wang et al. studied SL graphene using spatial light interference microscopy (a technique similar to that reported herein) but did not report any quantification of the optical properties mainly due to reconstruction artifacts that are inherent to this technique.<sup>29</sup>

In this article, we introduce a simple, fast, and accurate optical method based on high-resolution quantitative phase microscopy to image and determine the number of layers and characterize the optical properties of 2D materials. This technique uses a wavefront-sensing camera based on quadriwave lateral shearing interferometry (QLSI). This camera is mounted onto a lateral video port of an inverted transmission microscope to recover simultaneously the phase and intensity of an incoming light beam crossing the sample, using only one measurement. In the first part, we introduce the basic principle of QLSI. In the second part, we explain how QLSI measurements can be processed to retrieve the complex refractive index and the complex optical conductivity of 2D materials. The third and last part of the article focuses on experimental results. Results on single and multilayer graphene are shown to explain how the measured QLSI phase image can be used to (i) easily visualize transparent 2D materials, (ii) determine the number of layers of 2D materials, (iii) evidence the presence of a residual molecular layer, e.g., stemming from the fabrication process, and (iv) retrieve the complex refractive index and the complex optical conductivity of 2D materials. Results on  $MoS_2$  are subsequently presented to show the versatility of the technique and to explain that caution has to be used when working with 2D materials with a high refractive index.

## QUADRIWAVE LATERAL SHEARING INTERFEROMETRY

We introduce the basic principle of QLSI. We have chosen to use this phase and intensity measurement technique, but other techniques that quantitatively measure the scalar electromagnetic field in a nonbiased and halo-free manner could also be applied for investigation of the optical properties of 2D materials including, for example, digital holography and related techniques<sup>30–33</sup> or external reference-free approaches.<sup>34–36</sup>

Quadriwave lateral shearing interferometry is an optical imaging technique capable of mapping both the intensity and wavefront profiles of a light beam. A QLSI wavefront-sensing camera consists of a modified Hartmann grating (MHG) located just in front of a regular CCD camera.<sup>37</sup> The interferogram produced by the MHG and recorded by the CCD camera is subsequently processed to retrieve both the intensity and the wavefront profiles of the incoming light.<sup>38</sup> As this interferometric technique only involves a grating that is fixed onto a CCD sensor, it does not require complicated and sensitive alignments, like other interferometric techniques. The interferometric nature of the measurements makes this technique highly sensitive; QLSI can easily detect wavefront distortions smaller than 1 nm. Our QLSI camera (Sid4Bio from



**Figure 2.** Three descriptions to model the optical response of a 2D material. Model 1 considers the 2D material as an infinitely thin object characterized by a 2D optical conductivity  $\underline{\sigma}_{2D}$ . The transmission coefficient *t* is the ratio between the transmitted electric field  $E_t$  and the incident electric field  $E_i$ . Models 2 and 3 are equivalent. They are meant to model a 2D material as a slab of refractive index  $\underline{n}$ , and they take into account the occurrence of multiple reflections within the slab. In model 2, the transmission coefficient *t* is obtained as an infinite sum of terms stemming from multiple reflections ( $t_i$ ). The transmission coefficient *t* in model 3 is the ratio between the transmitted electric field  $E_t$  in the substrate and the incident electric field  $E_i$ .

Phasics SA) features a sensitivity of 0.3 nm·Hz<sup>-1/2</sup>. Note that this is not a limitation. The signal-to-noise ratio could be improved using more recent and sensitive cameras.<sup>39</sup>

Originally introduced to characterize laser beams, our group recently extended the scope of applications of QLSI by using QLSI wavefront-sensing cameras on inverted optical microscopes, where specific wavefront distortions are created by micrometric objects located at the sample plane. For instance, we have recently shown how QLSI cameras can be implemented on optical microscopes to image single living cells with high contrast<sup>40</sup> or microscale temperature profiles with a sensitivity of around 1 K.<sup>41</sup> The experimental configuration combining a QLSI camera and an optical microscope is depicted in Figure 1a. In this configuration, a Köhler illumination is used to illuminate the sample with a controlled optical plane wave (controlled illuminated area and controlled numerical aperture). In any measurement, a reference image is first taken over a clear area (without any object), prior to taking an image with the object of interest within the field of view. Let us name  $t_0$  the transmission of the sample without any object and t the transmission in the presence of the object of interest. We define the normalized complex transmission coefficient  $t/t_0$  as

$$\frac{t}{t_0} = \sqrt{T} e^{i\varphi} \tag{1}$$

where 
$$\varphi = \frac{2\pi}{\lambda} \delta l$$
 (2)

*T* is the transmittance of the object,  $\varphi$  is the phase retardation it induces, and  $\delta l$  is the wavefront distortion caused by the object, a physical quantity that is usually called the optical path difference (OPD). In this article, underlined letters, such as <u>*t*</u>, mean they are complex numbers. In this configuration, when considering the simplest case of an imaged object featuring a uniform refractive index *n*, embedded in a surrounding medium of refractive index *n*<sub>1</sub> as shown in Figure 1b, the OPD reads in first approximation (without taking into account multiple reflections within the slab)

$$\delta l(x, y) = (n - n_1) d(x, y) \tag{3}$$

This simple expression is widely used in quantitative phase imaging, for instance when studying biological samples.<sup>39,42</sup> However, we will explain in the next section that for some 2D materials this simple expression may be inaccurate.

#### QLSI APPLIED TO THE STUDY OF 2D MATERIALS

The scheme presented in the previous section suggests a straightforward use of QLSI for the study of 2D materials and the determination of their refractive index by using eq 3. However, this equation may not be appropriate in the case of 2D materials. Its validity depends mostly on the refractive index of the material and to a lesser extent on its thickness. For materials with large refractive indices, the multiple reflections inside the layer cannot be neglected anymore and the OPD is no longer as simple as in eq 3. For instance, eq 3 leads to an error in the determination of refractive index of 1% for graphene and 40% for  $MoS_2$ , as will be seen hereinafter. We explain in the following how this equation has to be refined to accurately apply to 2D materials.

Note that refractive indices, just like optical permittivities, are defined only for bulk materials. For this reason, the determination of the refractive index of a 2D material requires the consideration of a 2D material as a slab with a defined physical thickness. This can be relevant for thick 2D materials—such as multilayer graphene or  $MoS_2$ —but questionable for graphene. The thickness of a graphene layer is usually considered to be the interlayer distance of graphite. However, this choice is arbitrary since a SL has no well-defined boundaries (the electronic density is decaying exponentially from the atomic plane). Thus, the refractive index can only be defined as an *effective* quantity once an *effective* thickness of the material is chosen. For this reason, the refractive index of graphene will be noted  $n_{\text{eff}}$  in this article, corresponding to an assumed graphene thickness of  $d_{\text{eff}} = 0.335$  nm.

The fact that the refractive index is not a relevant quantity for 2D materials has already been noted in the literature.<sup>43</sup> This is the reason that another quantity is often considered: the two-dimensional optical conductivity  $\underline{\sigma}_{2D}$ . It is defined such that the 2D electronic current density  $\underline{J}_{2D}$  (charge per unit length and time) within the 2D materials reads

$$\underline{\mathbf{J}}_{2\mathrm{D}} = \underline{\boldsymbol{\sigma}}_{2\mathrm{D}} \underline{\mathbf{E}} \tag{4}$$

where  $\underline{E}$  is the electric field in the 2D material, in complex notation. This definition considers a normal incidence of the incoming light beam.

In the following, we explain how both the complex refractive index and the complex 2D optical conductivity can by measured by QLSI. Three models are described: the first model focuses on the complex 2D optical conductivity, and models 2 and 3 explain how the complex refractive index can be retrieved. Albeit equivalent, models 2 and 3 are both important to mention because they correspond to different physical pictures of the underlying physics.

**Model 1.** In the first model, we characterize the 2D material via its 2D optical conductivity  $\underline{\sigma}_{2D}$ . Here, the 2D material is considered as an infinitely thin layer with surface charges that interact with the incoming light. This picture is well adapted to the study of an atom-thick 2D material such as graphene. We consider that the 2D material is placed at the interface between two semi-infinite media represented by their refractive indices  $n_1$  and  $n_2$ . As presented in Figure 2, the propagation direction of the incoming light is normal to the interface and oriented from medium 1 (incident medium) to medium 2 (substrate). By using Maxwell's equations with the appropriate electromagnetic boundary conditions, one can obtain the complex transmission coefficient  $\underline{t}$  as a function of the complex 2D optical conductivity  $\underline{\sigma}_{2D}$  as follows:

$$\underline{t} = \frac{2n_1}{n_1 + n_2 + \frac{\sigma_{2D}}{\varepsilon_{0^c}}}$$
(5)

This expression is derived in the Supporting Information. By normalizing this quantity by the transmission coefficient in the absence of the 2D material  $t_0 = \frac{2n_1}{n_1 + n_2}$ , one obtains

$$\frac{\underline{t}}{t_0} = \frac{1}{1 + \frac{\underline{\sigma}_{2D}}{(n_1 + n_2)\varepsilon_0 c}}$$
(6)

Using eq 1, one can derive the expressions of the real  $\sigma_r$  and imaginary  $\sigma_i$  parts of  $\underline{\sigma}_{2D}$  as a function of the measured quantities by QLSI, *T*, and  $\delta l$ :

$$\sigma_{\rm r} = \varepsilon_0 c (n_1 + n_2) \left[ \frac{1}{\sqrt{T}} \cos\left(\frac{2\pi}{\lambda}\delta l\right) - 1 \right]$$
(7)

$$\sigma_{\rm i} = -\varepsilon_0 c(n_1 + n_2) \frac{1}{\sqrt{T}} \sin\left(\frac{2\pi}{\lambda}\delta l\right) \tag{8}$$

**Models 2 and 3.** We consider now the 2D material as a slab with a physical thickness *d* and characterized by a complex refractive index  $\underline{n} = n + i\kappa$ , where *n* is the refractive index and  $\kappa$  the extinction coefficient. To refine eq 3, we now take into account the multiple reflections of the incident light within the slab. For this purpose, two different models corresponding to two physical pictures can be considered (models 2 and 3 in Figure 2). Although these two models lead to exactly the same expression of the normalized transmission coefficient, we believe it is worth mentioning them both for the sake of comprehensiveness. Albeit equivalent, these two models correspond to totally different physical pictures that are both valuable.

Model 2 consists in writing the transmitted electric field as an infinite sum of terms stemming from the multiple reflections. A fraction of the transmitted light from medium 1 to the 2D material layer undergoes multiple reflections inside the layer. For each round-trip, a fraction of the light is transmitted to the third medium. The total transmission coefficient is obtained by the interference of all the transmitted beams to the third medium. The result of the calculation is given by eq 9, and the derivation of this expression is given in the Supporting Information.

Model 3 considers that each of the three media contains a planar electromagnetic wave (see Figure 2). In the first medium, the electromagnetic wave is decomposed into an incident and a reflected wave. The second medium (the 2D material layer) contains a transmitted and a reflected wave. The third medium contains only a transmitted wave. Using the Maxwell–Faraday equation for each medium and considering the appropriate electromagnetic boundary conditions, one can obtain the total transmission coefficient of the system (see eq 9). The derivation is given in the Supporting Information. Interestingly, this approach amounts to taking into account multiple reflections.

The expression of the complex transmission coefficient as a function of  $n_1$ ,  $n_2$ , and <u>n</u> obtained from models 2 and 3 reads

$$\underline{t} = \frac{4n_{\underline{n}}\underline{n}e^{ik_{0}d\underline{n}}}{(\underline{n}+n_{1})(\underline{n}+n_{2}) - (\underline{n}-n_{1})(\underline{n}-n_{2})e^{i2k_{0}d\underline{n}}}$$
(9)

where  $k_0 = \frac{2\pi}{\lambda}$  is the wavevector of the light in vacuum and *d* is the thickness of the 2D material. By normalizing this quantity by the transmission coefficient in the absence of the 2D material (i. e.,  $\underline{n} = n_1$ ), which reads  $\underline{t}_0 = \frac{2n_1}{n_1 + n_2} e^{ik_0 dn_1}$ , we get

$$\frac{\underline{t}}{\underline{t}_0} = \frac{2\underline{n}(n_1 + n_2)e^{ik_0d(\underline{n} - n_1)}}{(\underline{n} + n_1)(\underline{n} + n_2) - (\underline{n} - n_1)(\underline{n} - n_2)e^{i2k_0d\underline{n}}}$$
(10)

The complex quantity  $\underline{t}/\underline{t}_0$  can be determined by QLSI from the measurements of T and  $\delta l$ , following eq 1. Thus, the only unknown in eq 10 is  $\underline{n}$ , the physical quantity of interest. However, unlike eq 6, which can be easily inverted to express  $\underline{\sigma}_{2D}$ , eq 10 cannot be simply inverted to express  $\underline{n}$  as a function of  $\underline{t}/\underline{t}_0$ . Hence, eq 10 has to be solved numerically using an inversion algorithm. In our study, we used the function FindRoot of Wolfram Mathematica. The corresponding Mathematica notebook is provided in the Supporting Information.

Noteworthily, models 2 and 3 are relatively simple when dealing with two interfaces, which is the case in this work. However, these models become tedious when dealing with a larger number of interfaces, i.e., constituted by a stack of layers having different refractive indices. In this case, another model, named the transfer matrix formalism, is more suited to calculate the transmission coefficient of the system.<sup>44,45</sup>

#### EXPERIMENTAL RESULTS AND DISCUSSION

This section is devoted to the study of graphene and  $MoS_2$  by the QLSI technique. For clarity, we show in Figure 3 the atomic structures of graphene (right) and  $MoS_2$  (left). Graphene is



Figure 3. 3D scheme of the atomic structures of graphene (right) and  $MoS_2$  (left).



Figure 4. (a) OPD, (b) intensity images, and (c) their corresponding profiles for a particular graphene structure obtained by mechanical exfoliation and deposited on a glass substrate. The illumination source is centered at a wavelength of 625 nm. This observation was made with  $40 \times$  magnification and NA = 0.75. (d) Raman image for the same structure (integrated intensity of the G band).

composed of a single layer of carbon atoms.  $MoS_2$  is composed of a layer of molybdenum atoms sandwiched between two layers of sulfur atoms.

**Graphene.** Experimentally, we studied graphene structures fabricated by two different methods: mechanical exfoliation and chemical vapor deposition (CVD).

With exfoliated graphene, flakes are randomly scattered on a glass substrate, with thicknesses ranging from that of a single layer to a few tens of layers. As an illustrative example, we chose to study a particular graphene structure ideally composed of two different regions corresponding to SL and bilayer graphene, as supported by Raman measurement (see Figure 4d). Figure 4a and b show the OPD and intensity images of the graphene structure taken by QLSI, illuminated over a wavelength range of  $625 \pm 10$  nm. As shown in Figure 4c, the OPD for a SL graphene measured by QLSI is  $\delta l = 0.5 \pm 0.1$  nm. The error of 0.1 nm corresponds to the standard deviation of the background signal. In addition to the OPD, the transmittance of our sample can be determined from the normalized intensity image (see Figure 4b,c) defined as the intensity image divided by the reference intensity image. For SL graphene, we measured a transmittance of  $T = 98.5 \pm 0.1\%$ . The corresponding opacity is  $1 - T = 1.5 \pm 0.1\%$ . Note that this value is lower than the reported value for suspended graphene (substrate-free graphene), which is 2.3%,<sup>46</sup> because graphene opacity is also dependent on the refractive index of the surrounding medium,<sup>47</sup> as evidenced by eq 5.

Let us first consider a SL graphene as a slab with an effective thickness  $d_{\rm eff}$  = 0.335 nm. One can work out an effective

complex refractive index of graphene by inverting eq 10. Taking  $n_1 = 1$  (air) and  $n_2 = 1.50$  (glass substrate), it yields  $\underline{n}_{\text{eff}} = (2.46 \pm 0.16) + i(1.14 \pm 0.14)$ . If we use the simplistic expression of the OPD introduced in eq 3 to retrieve the real part of the refractive index, we get  $n = 2.49 \pm 0.3$ . One can see that in the case of graphene the error between the two approaches is only 1%. The reason that the simple eq 3 works well for graphene is that its refractive index is not too large. Figure 5 shows a comparison between the measured refractive index (n) and extinction coefficient  $(\kappa)$  in this work with those reported in



**Figure 5.** Complex refractive indices of SL graphene given in the literature and reported in this work at a wavelength close to 625 nm (Ni et al.,<sup>48</sup> Wang et al.,<sup>25</sup> Bruna and Borini,<sup>23</sup> Kravets et al.,<sup>49</sup> Wurstbauer et al.,<sup>27</sup> Matković et al.,<sup>52</sup> Ye et al.,<sup>50</sup> Cheon et al.<sup>51</sup>).

the literature at a wavelength close to 625 nm.<sup>23,25,27,48–51</sup> One can see that the effective complex refractive index determined by the QLSI technique is in general agreement with the literature. The variations from one value to another observed in this figure do not necessarily stem from systematic errors of the different techniques. Different graphene samples may have different doping levels, defects, or residues on top that may modify their optical properties.

As discussed in the previous section, characterizing graphene via its 2D optical conductivity instead of a refractive index makes more sense. By using eqs 7 and 8, the complex optical conductivity of a SL graphene at a wavelength of 625 nm is found to be  $\sigma_{2D} = (0.82 \pm 0.2)\sigma_0 - i(0.55 \pm 0.1)\sigma_0$ , where  $\sigma_0 = e^2/4\hbar$  is the universal conductivity in the limit of a massless Dirac–Fermion band structure. Table 1 lists measurements of

 Table 1. Comparison of the Complex Optical Conductivity

 of a SL Graphene Obtained in This Work to the Reported

 Values

reference	$\sigma_{ m r}~({ m in~unit~of}\ \sigma_{ m 0})$	$egin{array}{c} -\sigma_{ m i} \ ({ m in \ unit \ of} \ \sigma_0) \end{array}$	wavelength (nm)
this work	$0.82 \pm 0.2$	$0.55 \pm 0.1$	625
Chang et al. <sup>54</sup>	1.16	0.39	625
Wang et al. <sup>25</sup>	0.80	0.63	550
Ni et al. <sup>48</sup>	0.74	0.3	550
Bruna and Borini <sup>23</sup>	1	1.17	550
Mak et al. <sup>55</sup>	1.26		625
Gogoi et al. <sup>56</sup>	1.06		625

the optical conductivity of graphene reported in the literature. Most of the measurements in the visible range focused on the real part. Few studies measured the imaginary part. Note that Wang et al.,<sup>25</sup> Ni et al.,<sup>48</sup> and Bruna et al.<sup>23</sup> did not directly measure optical conductivities but permittivities of graphene. The conductivities of these studies mentioned in Table 1 were subsequently computed by Skulason et al.<sup>53</sup> from the values of their permittivities. The complex optical conductivity measured by QLSI is in general agreement with the literature. It may be surprising to observe that the measured values are so dispersed. As aforementioned, we believe this problem does not necessarily come from systematic errors related to the different experimental techniques. It can stem from the nature of the graphene samples, which may vary from one fabrication method to another. As will be shown in the next paragraph, some residue can remain on the sample and be difficult to detect. The Fermi level can also vary from one sample to another depending on the quality of the sample. For this reason, the values we report here have to be seen as specific values for the sample we observed, not as an attempt to measure a universal optical conductivity of graphene on glass. The study of the dispersion of graphene optical constants from one sample to another could form the basis of future studies.

We now present results on SL graphene samples fabricated by CVD.<sup>57–59</sup> These results are meant to show how QLSI can efficiently determine the presence and the nature of residue on graphene flakes. Experimentally, an extended SL graphene was grown on a copper substrate. The SL graphene has been subsequently transferred on a glass substrate by a wet chemical transfer process involving the use of a PMMA polymer layer, 300 nm thick, as a temporary graphene supporting layer. The polymer was then removed by acetone under heating.<sup>59</sup> The fabricated graphene layer has been eventually structured by O<sub>2</sub> plasma etching in order to get a 2D array of SL graphene disks, around 8  $\mu$ m in diameter. The phase and intensity images of this sample are shown in Figure 6a and b, respectively. With



**Figure 6.** (a) Phase and (b) intensity images of SL graphene fabricated by CVD technique and transferred on a glass substrate and then structured by plasma etching. The illumination source is centered at a wavelength of 625 nm. This observation was made with  $60 \times$  magnification and NA = 0.7. (c, d) Pixel-by-pixel histogram of the OPD and intensity images, respectively.

such a sample, the measured OPD of the SL graphene discs was about 2 nm (see Figure 6c). This OPD value is 4 times as large as that of exfoliated graphene, while the transmittance remains approximately the same (98.2%) as shown in the histogram of Figure 6d. The thicker measured OPD for SL CVD graphene suggests that this sample includes some other material in addition to graphene. The fact that some residual polymer layer can remain on graphene using this fabrication process is a wellknown problem,<sup>59</sup> but it is often difficult to evidence and to characterize. QLSI enables one to retrieve both the thickness  $d_{\rm I}$ and the refractive index  $n_{\rm L}$  of a possible residual layer. To retrieve these two quantities, it is sufficient to perform two QLSI measurements with two different surrounding media, such as air and water (i.e.,  $n_1 = 1$  and  $n_1 = 1.33$ ). Using eq 3 (a good approximation for graphene), one can derive a system of two equations where the two unknowns are  $n_{\rm L}$  and  $d_{\rm L}$  (see Supporting Information for the demonstration). We found  $n_{\rm L} =$ 1.4 and  $d_{\rm L} = 3.3$  nm. The thickness is consistent with AFM measurements,<sup>60</sup> and the refractive index is consistent with PMMA's. This example illustrates how QLSI can tell whether 2D materials obtained using wet transfer are clean and free from remaining chemical residues and highlights the difficulty to achieve complete removal of polymer used for the transfer process.

**Molybdenum Disulfide.** In order to demonstrate the versatility of QLSI and explain how QLSI can apply to thicker 2D materials, we conducted experiments on molybdenum disulfide ( $MoS_2$ ). It belongs to the family of transition metal dichalcogenides and does not consist of a single layer of atoms. It is thus thicker than graphene (see Figure 3). Figure 7a and b show the OPD and intensity images, respectively, for a SL



**Figure 7.** (a) Phase and (b) intensity images of SL MoS<sub>2</sub> fabricated by CVD on a glass substrate. The illumination source is centered at a wavelength of 625 nm. This observation was made with 100× magnification and NA = 1.3. (c) Comparison of the estimated effective complex refractive index in this work (refractive index and extinction coefficient of a SL MoS<sub>2</sub>) with those reported in the literature at a wavelength close to 625 nm (Liu et al.,<sup>62</sup> Morozov and Kuno,<sup>43</sup> Yu et al.,<sup>63</sup> Zhang et al.,<sup>64</sup> Mukherjee et al.,<sup>24</sup> Funke et al.<sup>28</sup>). (d) Complex optical conductivity of MoS<sub>2</sub> determined by our technique and compared to the values given by Morozov and Kuno.<sup>43</sup>

 $MoS_2$  flake fabricated by CVD and transferred on a glass substrate.<sup>61</sup> The transfer process of  $MoS_2$  on the glass substrate is described in the Supporting Information. These measurements are done with an illumination source centered at 625 nm. The obtained OPD and transmittance of SL  $MoS_2$  are 5.70  $\pm$  0.22 nm and 96.5  $\pm$  0.2% respectively. The corresponding effective complex refractive index obtained by inverting eq 10 is  $\underline{n}_{\rm eff} = (4.98 \pm 0.10) + i(0.66 \pm 0.06)$  using a thickness of SL  $MoS_2$  of  $d_{MoS_2} = 0.62$  nm. Figure 7c shows a comparison of the determined effective complex refractive index in this work with those reported in the literature at a wavelength close to 625 nm. One can see considerable variations in the refractive index and extinction coefficient obtained from the literature. This can be attributed to the fabrication process of the samples, which may modify the properties of  $MoS_2$ .

It is worth noting that, unlike graphene, eq 3 fails in the estimation of the refractive index of  $MoS_2$  by an error larger than 40% ( $n_{\rm eff} = 2.91 \pm 0.40$ ). This is due to the fact that the refractive index of  $MoS_2$  is higher than that of graphene, which makes the multiple reflections more efficient.

In addition to the complex refractive index, one can also estimate the complex optical conductivity of a SL MoS<sub>2</sub> by using eqs 7 and 8. This leads to a value of  $\underline{\sigma}_{2D} = [(1.40 \pm 0.35) - i(5.0 \pm 0.2)]2e^2/h$  at a wavelength close to 625 nm.

Unlike graphene,  $MoS_2$  is very dispersive in the visible range (i.e., its optical properties strongly depend on the wavelength).

We performed QLSI measurements at different wavelengths to investigate the optical properties of MoS<sub>2</sub> in the whole visible range. For this purpose, we have used a white light illumination source combined with bandpass filters, every 50 nm, with a bandwidth of 40 nm. As an example, we show in Figure 7d the real ( $\sigma_r$ ) and imaginary ( $\sigma_i$ ) parts of the optical conductivity of MoS<sub>2</sub> measured at different wavelengths. We also plotted in the same figure the complex optical conductivity estimated by Morozov and Kuno using transmission and reflection measurements.43 Comparisons of the complex refractive indices are shown in Supporting Information. Figure 7d shows a good agreement for the values of  $\sigma_r$ .  $\sigma_i$  has the same appearance but features lower values in comparison with those of Morozov and Kuno.<sup>43</sup> However, the MoS<sub>2</sub> samples used by Morozov and Kuno were fabricated by mechanical exfoliation, leading to a different quality of MoS<sub>2</sub> in comparison with CVD.

The studies reported above show that the QLSI is a powerful technique to characterize different 2D materials on transparent substrates. The only limitation of QLSI regarding the nature of the investigated 2D material is its transparency. Too thick materials may become opaque, and in this case one can no longer measure any wavefront distortion. However, we can overcome this limitation by working in the reflection geometry. This should be suited to image and characterize 2D materials in nontransparent samples.<sup>65</sup> This modality will be addressed in future work.

## **ACS Photonics**

# CONCLUSION

This article introduces a novel optical microscopy technique to characterize 2D materials. This method involves the use of a QLSI camera, which maps both the phase and the intensity of an incoming light beam, when implemented on a microscope. We show how such a simple scheme allows both the geometrical and the optical characterization for 2D materials like no other technique before. In particular, the main purpose of QLSI we evidenced is to determine the number of stacked layers and measure the complex optical conductivity and complex refractive index of 2D material structures.

The results presented in this article highlight different benefits of this technique compared to other characterization techniques of 2D materials. First, QLSI is noninvasive. For example, an irradiance of typically 100 mW·cm<sup>-2</sup> is used for QLSI measurements. This is 5 orders of magnitude lower than that used in Raman spectroscopy, where a laser of typically 1 mW is focused on the sample. Modifications of the optical properties of 2D materials have been evidenced after Raman characterization induced by the large intensity of the probe laser.<sup>14,15</sup> Then, compared with Raman spectroscopy, QLSI is fast. Single-layer graphene can be observed with an acquisition time of around 1 s, but it can be even faster using more advanced QLSI cameras.<sup>39</sup> These advantages make it possible to easily look for and visualize single-layer graphene, like with a regular camera. QLSI also enables a mapping of the optical properties, while other techniques such as transmittance or reflectance measurements rather perform ensemble measurement over spatially extended and uniform samples. For this reason, QLSI should be particularly powerful when applied to 2D material heterostacks. We have also shown how QLSI can detect and characterize the presence of an unwanted residual polymer thin film of a few nanometers on top of graphene, stemming from the fabrication process. Finally, QLSI allows for both morphological and optical characterizations, while other techniques usually achieve one or the other, which make QLSI particularly convenient and versatile.

For all these reasons, we expect QLSI to impact both the field of research working on 2D materials, by providing an effective and simple characterization technique, and the community working in quantitative phase imaging, by highlighting a new topic of interest for them: the study of 2D materials.

### ASSOCIATED CONTENT

### **S** Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.7b00845.

Derivation of the expressions of models 1, 2, and 3; additional results and details concerning the estimation of the residual layer on CVD-graphene; Raman spectrum of CVD-graphene sample; description of the transfer process of  $MoS_2$  sample; wavelength-dependent complex refractive index of  $MoS_2$  measured in this work and compared with the literature (PDF)

The mathematica notebook allowed the determination of the complex refractive index from models 2 and 3 (ZIP)

## AUTHOR INFORMATION

Corresponding Authors

\*E-mail: samira.khadir@Fresnel.fr.

\*E-mail: guillaume.baffou@Fresnel.fr. ORCID <sup>@</sup>

Samira Khadir: 0000-0002-2280-4099

Guillaume Baffou: 0000-0003-0488-1362

# Notes

The authors declare no competing financial interest.

#### ACKNOWLEDGMENTS

D.V. and E.G. acknowledge the financial support of the Renatech network. The authors thank J. R. Huntzinger for helpful discussions, in particular for pointing out that eq 3 was a priori not applicable for 2D materials. R. Parret and M. Paillet are also acknowledged for preparation and characterization of mechanically exfoliated graphene. N.M. acknowledges support from Science Foundation Ireland (15/SIRG/3329).

#### REFERENCES

(1) Novoselov, K. S.; Geim, A. K.; Morozov, S. V.; Jiang, D.; Katsnelson, M. I.; Grigorieva, I. V.; Dubonos, S. V.; Firsov, A. A. Twodimensional gas of massless Dirac fermions in graphene. *Nature* **2005**, 438, 197–200.

(2) Zhang, Y.; Tan, Y.; Stormer, H. L.; Kim, P. Experimental observation of the quantum Hall effect and Berry's phase in graphene. *Nature* **2005**, *438*, 201–204.

(3) Lee, C.; Wei, X.; Kysar, J. W.; Hone, J. Measurement of the Elastic Properties and Intrinsic Strength of Monolayer Graphene. *Science* **2008**, *321*, 385–388.

(4) Saha, S. K.; Baskey, M.; Majumdar, D. Graphene Quantum Sheets: A New Material for Spintronic Applications. *Adv. Mater.* **2010**, 22, 5531–5536.

(5) Kim, K.; Zhao, Y.; Jang, H.; Lee, S.; Kim, J.; Kim, K. S.; Ahn, J.; Kim, P.; Choi, J.; Hong, B. Large-scale pattern growth of graphene films for stretchable transparent electrodes. *Nature* **2009**, *457*, 706–710.

(6) Mishra, A.; Ramaprabhu, S. Functionalized Graphene-Based Nanocomposites for Supercapacitor Application. J. Phys. Chem. C 2011, 115, 14006–14013.

(7) Premkumar, T.; Geckeler, K. E. Graphene–DNA hybrid materials: Assembly, applications, and prospects. *Prog. Polym. Sci.* **2012**, *37*, 515–529.

(8) Arsat, R.; Breedon, M.; Shafiei, M.; Spizziri, P.; Gilje, S.; Kaner, R.; Kalantar-zadeh, K.; Wlodarski, W. Graphene-like nano-sheets for surface acoustic wave gas sensor applications. *Chem. Phys. Lett.* **2009**, 467, 344–347.

(9) Luo, Z.; Zhou, M.; Wu, D.; Ye, C.; Weng, J.; Dong, J.; Xu, H.; Cai, Z.; Chen, L. Graphene-Induced Nonlinear Four-Wave-Mixing and Its Application to Multiwavelength Q-Switched Rare-Earth-Doped Fiber Lasers. J. Lightwave Technol. **2011**, *29*, 2732–2739.

(10) Novoselov, K.; Geim, A.; Morozov, S.; Jiang, D.; Zhang, Y.; Dubonos, S.; Grigorieva, I.; Firsov, A. Electric Field Effect in Atomically Thin Carbon Films. *Science* **2004**, *306*, 666–669.

(11) Akinwande, D.; Petrone, N.; Hone, J. Nat. Commun. 2014, 5, 5678.

(12) Shavanova, K.; Bakakina, Y.; Burkova, I.; Shtepliuk, I.; Viter, R.; Ubelis, A.; Beni, V.; Starodub, N.; Yakimova, R.; Khranovskyy, V. Application of 2D Non-Graphene Materials and 2D Oxide Nanostructures for Biosensing Technology. *Sensors* **2016**, *16*, 223.

(13) Gupta, A.; Chen, G.; Joshi, P.; Tadigadapa, S.; Eklund. Raman Scattering from High-Frequency Phonons in Supported n-Graphene Layer Films. *Nano Lett.* **2006**, *6*, 2667–2673.

(14) Tiberj, A.; Rubio-Roy, M.; Paillet, M.; Huntzinger, J. R.; Landois, P.; Mikolasek, M.; Contreras, S.; Sauvajol, J. L.; Dujardin, E.; Zahab, A. A. Reversible optical doping of graphene. *Sci. Rep.* **2013**, *3*, 2355.

(15) Krauss, B.; Lohmann, T.; Chae, D.-H.; Haluska, M.; von Klitzing, K.; Smet, J. H. Laser-induced disassembly of a graphene single

crystal into a nanocrystalline network. Phys. Rev. B: Condens. Matter Mater. Phys. 2009, 79, 165428.

(16) 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.

(17) Roddaro, S.; Pingue, P.; Piazza, V.; Pellegrini, V.; Beltram, F. The Optical Visibility of Graphene: Interference Colors of Ultrathin Graphite on SiO2. *Nano Lett.* **2007**, *7*, 2707–2710.

(18) Blake, P.; Hill, E. W.; Castro Neto, A. H.; Novoselov, K. S.; Jiang, D.; Yang, R.; Booth, T. J.; Geim, A. K. Making graphene visible. *Appl. Phys. Lett.* **200**7, *91*, 063124.

(19) Ouyang, W.; Liu, X.-Z.; Li, Q.; Zhang, Y.; Yang, J.; shui Zheng, Q. Optical methods for determining thicknesses of few-layer graphene flakes. *Nanotechnology* **2013**, *24*, 505701.

(20) Gao, L.; Ren, W.; Li, F.; Cheng, H. Total Color Difference for Rapid and Accurate Identification of Graphene. *ACS Nano* **2008**, *2*, 1625–1633.

(21) Wang, Y.; Gao, R.; Ni, Z.; He, H.; Guo, S.; Yang, H.; Cong, C.; Yu, T. Thickness identification of two-dimensional materials by optical imaging. *Nanotechnology* **2012**, *23*, 495713.

(22) Cheon, S.; Kihm, K.; Park, J.; Lee, J.; Lee, B.; Kim, H.; Hong, B. How to optically count graphene layers. *Opt. Lett.* **2012**, *37*, 3765.

(23) Bruna, M.; Borini, S. Optical constants of graphene layers in the visible range. *Appl. Phys. Lett.* **2009**, *94*, 031901.

(24) Mukherjee, B.; Tseng, F.; Gunlycke, D.; Amara, K.; Eda, G.; Simsek, E. Complex electrical permittivity of the monolayer molybdenum disulfide ( $MoS_2$ ) in near UV and visible. *Opt. Mater. Express* **2015**, *5*, 447–455.

(25) Wang, X.; Chen, Y. P.; Nolte, D. D. Strong anomalous optical dispersion of graphene: complex refractive index measured by Picometrology. *Opt. Express* **2008**, *16*, 22105.

(26) Efraín, O.; Gabás, M.; Barrutia, L.; Pesquera, A.; Centeno, A.; Palanco, S.; Zurutuza, A.; Algora, C. Determination of a refractive index and an extinction coefficient of standard production of CVD-graphene. *Nanoscale* **2014**, *7*, 1491–1500.

(27) Wurstbauer, U.; Röling, C.; Wurstbauer, U.; Wegscheider, W.; Vaupel, M.; Thiesen, P. H.; Weiss, D. Imaging ellipsometry of graphene. *Appl. Phys. Lett.* **2010**, *97*, 231901.

(28) Funke, S.; Miller, B.; Parzinger, E.; Thiesen, P.; Holleitner, A.; Wurstbauer, U. Imaging spectroscopic ellipsometry of MoS2. *J. Phys.: Condens. Matter* **2016**, *28*, 385301.

(29) Wang, Z.; Chun, I. S.; Li, X.; Ong, Z. Y.; Pop, E.; Millet, L.; Gilette, M.; Popescu, G. Topography and Refractometry of Nanostructures Using Spatial Light Interference Microscopy. *Opt. Lett.* **2010**, *35*, 208–210.

(30) Cuche, E.; Bevilacqua, F.; Depeursinge, C. Digital holography for quantitative phase-contrast imaging. *Opt. Lett.* **1999**, *24*, 291–293.

(31) Ikeda, T.; Popescu, G.; Dasari, R. R.; Feld, M. S. Hilbert phase microscopy for investigating fast dynamics in transparent systems. *Opt. Lett.* **2005**, *30*, 1165–1167.

(32) Kemper, B.; von Bally, G. Digital holographic microscopy for live cell applications and technical inspection. *Appl. Opt.* **2008**, *47*, A52–A61.

(33) Shang, R.; Chen, S.; Li, C.; Zhu, Y. Spectral modulation interferometry for quantitative phase imaging. *Biomed. Opt. Express* **2015**, *6*, 473–479.

(34) Parthasarathy, A. B.; Chu, K. K.; Ford, T. N.; Mertz, J. Quantitative phase imaging using a partitioned detection aperture. *Opt. Lett.* **2012**, *37*, 4062–4064.

(35) Lee, K.; Park, Y. Quantitative phase imaging unit. *Opt. Lett.* **2014**, *39*, 3630–3633.

(36) Waller, L.; Tian, L.; Barbastathis, G. Transport of Intensity phase-amplitude imaging with higher order intensity derivatives. *Opt. Express* **2010**, *18*, 12552–12561.

(37) Primot, J.; Guérineau, N. Extended Hartmann Test Based on the Pseudoguiding Property of a Hartmann Mask Completed by a Phase Chessboard. *Appl. Opt.* **2000**, *39*, 5715–5720. (38) Bon, P.; Monneret, S.; Wattellier, B. Noniterative Boundary-Artifact-Free Wavefront Reconstruction from its Derivatives. *Appl. Opt.* **2012**, *51*, 5698–5704.

(39) Bon, P.; Lécart, S.; Fort, E.; Lévêque-Fort, S. Fast Label-Free Cytoskeletal Network Imaging in Living Mammalian Cells. *Biophys. J.* **2014**, *106*, 1588–1595.

(40) Bon, P.; Maucort, G.; Wattellier, B.; Monneret, S. Quadriwave lateral shearing interferometry for quantitative phase microscopy of living cells. *Opt. Express* **2009**, *17*, 13080–94.

(41) Baffou, G.; Bon, P.; Savatier, J.; Polleux, J.; Zhu, M.; Merlin, M.; Rigneault, H.; Monneret, S. Thermal Imaging of Nanostructures by Quantitative Optical Phase Analysis. *ACS Nano* **2012**, *6*, 2452–2458.

(42) Wolf, E. Three-dimensional structure determination of semitransparent objects from holographic data. *Opt. Commun.* 1969, 1, 153–156.

(43) Morozov, Y.; Kuno, M. Optical constants and dynamic conductivities of single layer MoS2, MoSe2, and WSe2. *Appl. Phys. Lett.* **2015**, *107*, 083103.

(44) MacLeod, H. A. Thin-Film Optical Filters, 4th ed.; CRC Press, 2010.

(45) Heavens, O. S. Optical properties of thin films. *Rep. Prog. Phys.* **1960**, 23, 1.

(46) Nair, R.; Blake, P.; Grigorenko, A.; Novoselov, K.; Booth, T.; Stauber, T.; Peres, N.; Geim, A. Fine Structure Constant Defines Visual Transparency of Graphene. *Science* **2008**, *320*, 1308–1308.

(47) Jang, C.; Adam, S.; Chen, J.; Williams, E.; Sarma, S.; Fuhrer, M. Tuning the Effective Fine Structure Constant in Graphene: Opposing Effects of Dielectric Screening on Short- and Long-Range Potential Scattering. *Phys. Rev. Lett.* **2008**, *101*, 146805.

(48) Ni, Z.; Wang, H.; Kasim, J.; Fan, H.; Yu, T.; Wu, Y.; Feng, Y.; Shen, Z. Graphene Thickness Determination Using Reflection and Contrast Spectroscopy. *Nano Lett.* **2007**, *7*, 2758–63.

(49) Kravets, V.; Grigorenko, A.; Nair, R.; Blake, P.; Anissimova, S.; Novoselov, K.; Geim, A. Spectroscopic ellipsometry of graphene and an exciton-shifted van Hove peak in absorption. *Phys. Rev. B: Condens. Matter Mater. Phys.* **2010**, *81*, 155413.

(50) Ye, Q.; Wang, J.; Liu, Z.; Deng, Z.; Kong, X.; Xing, F.; Chen, X.; Zhou, W.; Zhang, C.; Tian, J. Polarization-dependent optical absorption of graphene under total internal reflection. *Appl. Phys. Lett.* **2013**, *102*, 021912.

(51) Cheon, S.; Kihm, K.; Kim, H.; Lim, G.; Park, J.; Lee, J. How to Reliably Determine the Complex Refractive Index (RI) of Graphene by Using Two Independent Measurement Constraints. *Sci. Rep.* **2014**, *4*, 6364.

(52) Matković, A.; Beltaos, A.; Milićević, M.; Ralević, U.; Vasić, B.; Jovanović, D.; Gajić, R. Spectroscopic imaging ellipsometry and Fano resonance modeling of graphene. J. Appl. Phys. **2012**, 112, 123523.

(53) Skulason, H.; Gaskell, P.; Szkopek, T. Optical Reflection and Transmission Properties of Exfoliated Graphite from a Graphene Monolayer to Several Hundred Graphene Layers. *Nanotechnology* **2010**, *21*, 295709.

(54) Chang, Y.; Liu, C.; Liu, C.; Zhong, Z.; Norris, T. Extracting the complex optical conductivity of mono- and bilayer graphene by ellipsometry. *Appl. Phys. Lett.* **2014**, *104*, 261909.

(55) Mak, K.; Shan, J.; Heinz, T. Seeing Many-Body Effects in Singleand Few-Layer Graphene: Observation of Two-Dimensional Saddle-Point Excitons. *Phys. Rev. Lett.* **2011**, *106*, 046401.

(56) Gogoi, P.; Santoso, I.; Saha, S.; Wang, S.; Neto, A.; Loh, K.; Venkatesan, T.; Rusydi, A. Optical conductivity study of screening of many-body effects in graphene interfaces. *Epl Europhys. Lett.* **2012**, *99*, 67009.

(57) Li, X.; Cai, W.; An, J.; Kim, S.; Nah, J.; Yang, D.; Piner, R.; Velamakanni, A.; Jung, I.; Tutuc, E.; Banerjee, S. K.; Colombo, L.; Ruoff, R. S. Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils. *Science* **2009**, *324*, 1312–1314.

(58) Reina, A.; Jia, X.; Ho, J.; Nezich, D.; Son, H.; Bulovic, V.; Dresselhaus, M. S.; Kong, J. Large Area, Few-Layer Graphene Films on Arbitrary Substrates by Chemical Vapor Deposition. *Nano Lett.* **2009**, *9*, 30–35.

(59) Deokar, G.; Avila, J.; I, R.; Codron, J.; Boyaval, C.; Galopin, E.; Asensio, M.; Vignaud, D. Towards high quality CVD graphene growth and transfer. *Carbon* **2015**, *89*, 82–92.

(60) Liu, L.; Zhou, H.; Cheng, R.; Yu, W. J.; Liu, Y.; Chen, Y.; Shaw, J.; Zhong, X.; Huang, Y.; Duan, X. High-Yield Chemical Vapor Deposition Growth of High-Quality Large-Area AB-Stacked Bilayer Graphene. *ACS Nano* **2012**, *6*, 8241–8249.

(61) O'Brien, M.; McEvoy, N.; Hanlon, D.; Hallam, T.; Coleman, J. N.; Duesberg, G. S. Mapping of Low-Frequency Raman Modes in CVD-Grown Transition Metal Dichalcogenides: Layer Number, Stacking Orientation and Resonant Effects. *Sci. Rep.* **2016**, *6*, 19476.

(62) Liu, H.; Shen, C.; Su, S.; Hsu, C.; Li, M.; Li, L. Optical properties of monolayer transition metal dichalcogenides probed by spectroscopic ellipsometry. *Appl. Phys. Lett.* **2014**, *105*, 201905.

(63) Yu, Y.; Yu, Y.; Cai, Y.; Li, W.; Gurarslan, A.; Peelaers, H.; Aspnes, D. E.; de Walle, C. G.; Nguyen, N. V.; Zhang, Y.; Cao, L. Exciton-dominated Dielectric Function of Atomically Thin MoS2 Films. *Sci. Rep.* **2015**, *5*, 16996.

(64) Zhang, H.; Ma, Y.; Wan, Y.; Rong, X.; Xie, Z.; Wang, W.; Dai, L. Measuring the Refractive Index of Highly Crystalline Monolayer MoS2 with High Confidence. *Sci. Rep.* **2015**, *5*, 8440.

(65) Bon, P.; Belaid, N.; Lagrange, D.; Bergaud, C.; Rigneault, H.; Monneret, S.; Baffou, G. Three-dimensional temperature imaging around a gold microwire. *Appl. Phys. Lett.* **2013**, *102*, 244103.