Visibility of graphene flakes on a dielectric substrate

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We model the optical visibility of monolayer and bilayer graphene deposited on a SiO$_2$/Si substrate or thermally annealed on the surface of SiC. Visibility is much stronger in reflection than in transmission, reaching the optimum conditions when the bare substrate transmits light resonantly. In the optical range of frequencies a bilayer is approximately twice as visible as a monolayer thereby making the two types of graphene distinguishable from each other.

Monolayer graphene is a single two-dimensional honeycomb lattice of carbon atoms. Although the first graphene-based structures were only recently fabricated [1] they have quickly become the subject of an extensive research effort [2, 3, 4]. Monolayer graphene is a zero-gap semiconductor with a Dirac-like dispersion of chiral quasiparticles near the $K$ points of the hexagonal first Brillouin zone [5]. Bilayer graphene is a pair of graphene sheets with the Bernal (AB) stacking arrangement. In the low-energy spectrum of this material [6] the conduction and valence bands both consist of two quadratic branches split by the inter-layer coupling $\gamma_1$. Measurements of the quantum Hall effect [1, 2, 7] and ARPES experiments [8] have confirmed that these are the low-energy band structures of these materials.

The widespread microcleavage technique used to fabricate graphene-based devices requires a visual inspection of the substrate [1] to find flakes of one or two layers thickness. In this Letter, we aim to determine the optimum conditions for making these flakes optically visible when they are deposited on various substrates. The parameters at one’s disposal (see Fig. 1) are the frequency $\omega$, angle $\alpha$ and aperture $\delta\alpha$ of the focused incident radiation, as well as the thicknesses of the various layers of the underlying dielectric materials.

Below we calculate the reflection of non-polarized incident light taking the geometry of the substrate into account with suitable boundary conditions at each of the interfaces between materials, appropriate frequency-dependent dielectric functions $\varepsilon(\omega)$ for each layer, and $\mu = 1$. Throughout the calculation, we use the data [9] available in the existing literature for the dispersion of the permittivity of silicon [10], silicon oxide [11] and silicon carbide [12]. With reference to Fig. 1, we analyze the reflection $R$ of light from a substrate with a flake on it and compare this to the reflection $R_0$ of a bare (graphene-free) substrate. The optical visibility of a flake is then determined as the contrast between two such parts of the sample studied using a monochromatic light source:

$$V_R = (R - R_0)/R_0.$$  \hspace{1cm} (1)

The scattering of light is analyzed using the electromagnetic wave equations in vacuum and dielectric media and the standard boundary conditions at interfaces between different materials,

$$\vec{E}_1^\parallel = \vec{E}_2^\parallel, \quad \vec{D}_1^\perp = \vec{D}_2^\perp, \quad \vec{B}_1^\parallel - \vec{B}_2^\parallel = \sigma(\omega)\vec{E} \times \vec{n}. \hspace{1cm} (2)$$

The superscripts $\parallel$ and $\perp$ stand for the components of the field parallel and perpendicular to the interface respectively, $\vec{n}$ is the unit vector normal to the interface, the subscript 1 (2) denotes the field below (above) the interface, and $\sigma(\omega)$ is the frequency-dependent conductivity of a graphene flake and $\vec{D} = \epsilon(\omega)\vec{E}$. One more boundary condition (on the perpendicular components of $\vec{H}$) duplicates Snell’s law.

Having in mind an optical setup used to locate a small flake, we consider a beam of light focused by a lens, so that the light in the beam arrives at the substrate surface with some aperture $\delta\alpha$ (see Fig. 1). Therefore the measurable reflectance to be used in Eq. (1) is

$$R(\alpha, \delta\alpha) = \int d\Omega \frac{1}{R(\vec{k})} P(\vec{k}), \hspace{1cm} (3)$$

where $P(\vec{k})$ characterises the spread of the beam over the solid angle of the aperture $\delta\alpha$ around $\alpha$, $\vec{k} = \vec{\alpha} (\sin \alpha, 0, -\cos \alpha)$ is the wave vector of the incident ray of light, and $R(\vec{k})$ is the reflection coefficient for a plane wave with this wave vector. Below we assume that the beam is equally dense at all angles within an aperture of $\delta\alpha$ around $\alpha$.

To describe the conductivity of graphene, we follow the method used in Refs [13, 14] taking into account the split
bands formed in the bilayer [3]. At low temperatures the result for the monolayer which takes into account the transition between the valence and conduction bands in the Dirac spectrum is \( \sigma_1 = e^2/4h \) (with a negligible imaginary part) [13]. This corresponds [13] to the absorption coefficient \( g = 4\pi\sigma/c \) which gives \( \sigma_1 = \pi e^2/hc \approx 2.5\% \).

For the bilayer, there are four possible inter-band transitions, reflected by its conductivity,

\[
\sigma_2 = \frac{e^2}{2h} \left( \frac{\Omega + 2}{2\Omega + 1} + \frac{1}{\Omega^2} (\Omega - 1) + \frac{1}{2\Omega - 1} \right) \\
+ i \frac{e^2}{2\pi h} \left( \frac{\Omega}{\Omega^2 - 1} \log \Omega + \frac{2}{\Omega} \log \left( \frac{1 + \Omega}{1 - \Omega} \right) \right) \\
- \frac{1}{2} \log \left( \frac{2 + \Omega}{2 - \Omega} \right) - \frac{1}{2} \log \left( 4 - \Omega^2 \right) \right) \tag{4}
\]

Here \( \Omega = \hbar \omega/\gamma_1 \) is the frequency written in units of the inter-layer coupling and \( \theta(x) = (1 + \text{sgn}(x))/2 \). The real part of this function has a discontinuity at \( \hbar \omega = \gamma_1 \approx 0.4\text{eV} \) and a cusp at \( \hbar \omega = 2\gamma_1 \). These correspond to the activation (at zero temperature) of the interband transitions between low-energy bands and split band, and the two split bands respectively. The imaginary part of \( \sigma_2 \) shows a divergence at \( \hbar \omega = \gamma_1 \), leading to an enhanced reflectance of the bilayer at this frequency.

For non-polarized light arriving at the incidence angle \( \alpha \) to the sample depicted on the right-hand side of Fig. 1 with graphene deposited on the top surface, the reflectance is

\[
\begin{align*}
R &= \frac{1}{2} \left| \sqrt{\varepsilon_s} \cos \alpha D - (\cos \alpha - \frac{4\pi \sigma}{\varepsilon_s}) C \right|^2 \\
+ \frac{1}{2} \left| \sqrt{\varepsilon_s} \cos \alpha C' - \cos \alpha D' \left( \frac{1 - \frac{4\pi \sigma}{\varepsilon_s \cos \alpha}}{\frac{1 + \frac{4\pi \sigma}{\varepsilon_s \cos \alpha}}{\varepsilon_s \cos \alpha} \cos \alpha} \right) \right|^2 ; \tag{5}
\end{align*}
\]

In this result the first term represents reflection of radiation polarized so that the electric field is perpendicular to the plane of incidence, the second term to radiation polarised so that the electric field is parallel to the plane of incidence, and

\[
\begin{align*}
A &= -\sqrt{\varepsilon_d} \cos \alpha \cos X_d + i \sqrt{\varepsilon_d} \cos \alpha \sin X_d, \\
B &= i \sqrt{\varepsilon_d} \cos \alpha \cos X_d - \sqrt{\varepsilon_s} \cos \alpha \cos X_d, \\
C &= -i \sqrt{\varepsilon_d} \cos \alpha \cos X_d + i \sqrt{\varepsilon_s} \cos \alpha \cos X_d, \\
D &= \sqrt{\varepsilon_d} \cos \alpha \cos X_d - i \sqrt{\varepsilon_s} \cos \alpha \cos X_d;
\end{align*}
\]

\[
\begin{align*}
A' &= \sqrt{\varepsilon_d} \cos \alpha \cos X_d - i \sqrt{\varepsilon_d} \cos \alpha \sin X_d, \\
B' &= i \sqrt{\varepsilon_d} \cos \alpha \cos X_d - \sqrt{\varepsilon_s} \cos \alpha \cos X_d, \\
C' &= i \sqrt{\varepsilon_d} \cos \alpha \cos X_d - i \sqrt{\varepsilon_s} \cos \alpha \cos X_d, \\
D' &= -i \sqrt{\varepsilon_d} \cos \alpha \cos X_d + \sqrt{\varepsilon_s} \cos \alpha \cos X_d.
\end{align*}
\]

Here \( X_s = \sqrt{\varepsilon_s} k_s \cos \alpha, \quad X_d = \sqrt{\varepsilon_d} k_d \cos \alpha, \quad \sin \alpha_b = \sin \alpha/\sqrt{\varepsilon_s}, \quad \sin \alpha_s = \sin \alpha/\sqrt{\varepsilon_d}, \quad \sin \alpha_d = \sin \alpha / \sqrt{\varepsilon_d}. \) The angle \( \alpha \) is determined by the direction of the wave vector of the incident plane wave, see Fig. 1. To model a finite slab of silicon of width \( d \) with a silicon oxide layer of width \( s \) on top, we substitute \( \varepsilon_d = \varepsilon_d(\omega), \quad \varepsilon_s = \varepsilon_s(\omega), \quad \varepsilon_b = 1, \) and the quantity \( R_0 \) is found by replacing \( \sigma = 0 \) in these expressions. To evaluate the visibility \( V_R \), the integral in Eq. (3) must be taken for \( R \) and \( R_0 \) using Eq. (5).

Figure 2 illustrates the visibility of mono- and bilayer flakes on a Si substrate of widths 0.5\( \mu \)\text{m} < \( d \) < 1.5\( \mu \)\text{m} and a 300nm SiO\text{2} layer (see Fig. 1) for light with 0.3eV < \( \hbar \omega < 2.5\text{eV} \) arriving with aperture \( \delta \alpha = 10^\circ \) around \( \alpha = 20^\circ \). The rapid oscillations of the visibility in this plot are caused by the resonant condition of the Si layer. When this layer is strongly transmitting (that is, when \( \cos X_d \approx 0 \)), the visibility is at its highest. This fine structure is modulated by the corresponding resonance condition in the oxide which is responsible for the ‘bands’ which lie across the plots in Fig. 2. The condition for maximum transmission through the oxide is \( \cos X_s \approx 0 \) which leads to

\[
\omega \approx c(n + \frac{1}{2})\pi/(s\sqrt{\varepsilon_s \cos \alpha_s}) \tag{6}
\]

where \( n \) is an integer. The wave vector of the light in the slab is of the order of an inverse micron, so the resonant conditions are closely spaced on the length scale of the substrate thickness. The visibility of a bilayer flake is higher than the visibility of a monolayer for \( \hbar \omega > \gamma_1 \approx 0.4\text{eV} \) because the conductivity of the bilayer is essentially twice as large as the conductivity of the monolayer in this energy range. Additionally, the divergence in the imaginary part of the bilayer conductivity at \( \hbar \omega = \gamma_1 \approx 0.4\text{eV} \) causes a stronger reflection and hence a larger visibility. Also we have calculated the transmittance \( T \) of the sample, and the corresponding visibility \( V_T = (T - T_0)/T_0 \) is shown in Fig. 2(b) where the same resonant structure appears, but is at least ten
times weaker than the visibility in reflectance.

We find that the visibility of graphene in reflectance is further enhanced by using a thick (semi-infinite) substrate with a sizeable oxide layer on its surface, in agreement with a recent experimental observation [16]. Figure 3(a) shows the visibility of graphene deposited on a semi-infinite slab of silicon [9] with a 300nm SiO$_2$ layer. In this case the analytical expression for the reflectance of a plane wave with wave vector $\vec{k} = \frac{\omega}{c} (\sin \alpha, 0, -\cos \alpha)$ can be found by substituting $\varepsilon_b = \varepsilon_{\text{Si}}(\omega)$, $\varepsilon_s = \varepsilon_{\text{SiO}_2}$ and $d = 0$ into Eq. (3). As before, both $R_b(k)$ and $R_0(k)$ (which is determined from this equation with $\sigma = 0$), must be substituted in Eq. (3) before the visibility is evaluated. In the plots in Fig. 3(a), the main features are the very strong reflectance of the graphene flake at $\hbar \omega \approx 0.5$eV and $\hbar \omega \approx 1.6$eV. These are due to the standing wave resonances in the oxide layer at the condition in Eq. (6). In Fig. 3(a) the peak in visibility at $\hbar \omega \approx 0.5$eV, $(n = 0)$ corresponds to the first resonance in the oxide layer and the peak at $\hbar \omega \approx 1.6$eV $(n = 1)$ to the second resonance. The factor of 2 difference between the bilayer and monolayer conductivities at $\hbar \omega \gg \gamma_1$ and the divergence in the imaginary part of $\sigma_2(\omega)$ at $\hbar \omega = \gamma_1 \approx 0.3$eV are manifest in the visibility.

Besides being produced using the microcleavage technique, ultra-thin graphitic films can also be grown by thermal annealing of SiC wafers [3] [17]. The reflectance for this configuration can be found by substituting $\varepsilon_2 = 0$, $\varepsilon_b = 1$ and $\varepsilon_s = \varepsilon_{\text{SiC}}$ in Eq. (5). Plots of the visibility defined by this function are shown in Fig. 3(b). The standing wave resonance in the substrate is again the main factor for the visibility of graphene, though it is weaker for a SiC slab than for the SiO$_2$/Si substrates.

In conclusion, we have found that graphene is much more visible in reflection than in transmission and that the resonance condition of the substrate is the dominating factor in determining its visibility. For optimum visibility the wavelength of monochromatic light used should be selected using Eq. (6), and for the visible frequency range where $(\sigma_2 \approx 2\sigma_1)$ a bilayer is clearly distinguishable from a monolayer.

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