Supporting Online Material for

Fine Structure Constant Defines Visual Transparency of Graphene
R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, A. K. Geim*
*To whom correspondence should be addressed. E-mail: geim@man.ac.uk

Published 3 April 2008 on Science Express
DOI: 10.1126/science.1156965

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MATERIALS AND METHODS

Fabrication of graphene membranes
Large graphene crystals were prepared by micromechanical cleavage of natural graphite ([www.graphit.de](http://www.graphit.de)) on top of an oxidized Si wafer ([S1](#)) with an additional thin layer of PMMA ([S2](#)). The latter significantly improved adhesion and allowed us to make graphene monocrystals that could easily exceed 100 μm in size. We used NITTO tape to minimize contamination by adhesive residues. Single-, double- or few-layer crystallites were identified in an optical microscope due to their different contrast that increases with increasing the number of layers ([S2](#)). The number of layers was also verified with atomic force and Raman microscopy.

By using photolithography, a perforated 20-μm-thick copper-gold film was deposited on top of the found crystallites. The films usually had 9 small apertures with diameters 20, 30 and 50 μm (see inset of Fig. 1A and [Fig. S1](#)), and the graphene crystallites were aligned against the apertures to cover them completely or partially. The Cu/Au film also served as a support structure (scaffold) and was 3 mm in diameter so that it could be used in standard holders for transmission electron microscopy (TEM). At the final stage of microfabrication, the scaffold was lifted off by dissolving the sacrificial PMMA layer, which left graphene attached to the scaffold (the use of a critical point dryer was essential).

The resulting devices could easily be handled and transferred between different measurement facilities. The developed technique allows a reliable and routine fabrication of practically macroscopic graphene membranes suitable for optical, electron-microscopy or other studies (our success rate in making the final devices is >50%). This is a significant technological advance with respect to the earlier fabrication procedures that largely relied on chance and allowed graphene membranes of only a few microns in size ([S3](#), [S4](#)).

Optical measurements
To measure the optical spectra, we used a xenon lamp (250-1200nm) as a light source and focused its beam on graphene membranes. The transmitted light intensity was measured by Ocean Optics HR2000 spectrometer. The recorded signal was then compared with the one obtained by directing the light beam through either an empty space or, as a double check, another aperture of the same size but without graphene. Typical experimental data are shown in Figure S2 by open circles. Here, to reduce the measurement noise below 0.1%, we have averaged the spectral curves over intervals Δλ of 10 nm.

An interesting alternative method to measure optical spectra of graphene was to use membranes that only partially covered the apertures (such as shown in Fig. S1) and take their images in an optical microscope (we used Nikon Eclipse LV100) using 22 different narrow-band-pass filters for transmission illumination. The images taken by high-quality grey and color cameras (Nikon DS2MBW and DS2Mv) were then analyzed, and
relative intensities of the light transmitted through different areas were calculated. Examples of such spectroscopy for graphene and its bilayer are shown in Fig. S3. Results of the two measurement techniques are compared in Figure S2 (circles versus squares) and show nearly the same accuracy. Note that the use of an optical microscope is possible for graphene membranes because they mostly absorb light with only a minute portion of it being reflected (<0.1%). The latter ensures that the opacity of graphene is practically independent of the numerical aperture and magnification (this was carefully checked experimentally and is in agreement with our calculations).

Both approaches to measure graphene transmittance spectra show a deviation from a constant opacity for $\lambda < 500$ nm (photon energy $E > 2.5$eV), and the same is valid for bilayer graphene (see Fig. 1B, S2 and S3). Such rapid deviations are not expected in theory (see below). We have investigated this spectral feature further and found that it is connected with surface contamination of graphene membranes by hydrocarbons. Graphene is extremely lipophilic and hydrocarbon contamination is practically impossible to avoid for samples exposed to air (a hydrocarbon layer partially covering graphene is always found in TEM; see, for example, ref. (S3)). To this end, we annealed our membranes in a hydrogen-argon atmosphere (S5) at 200°C, which significantly improved their cleanliness, as observed in TEM by using the membranes immediately after their annealing. The annealing is also found to significantly weaken the downturn in the violet-light transmittance but did not affect the spectra for $\lambda > 500$nm, which indicates that hydrocarbons are indeed responsible for this additional opacity (or, at least, most of it). Here we note that many polymer (hydrocarbon) materials, especially those used in lithography, have an absorption edge in violet light. Alternatively, we speculate that it could be a tail of the plasmon resonance expected at $E \approx 5$eV, which is broadened by surface contaminants.

**SUPPLEMENTARY TEXT**

**Universal dynamic conductivity of graphene**

Optical properties of thin films are commonly described in terms of dynamic or optical conductivity $G$. For a two-dimensional (2D) Dirac spectrum with a conical dispersion relation $\varepsilon = \hbar v_F |k|$ ($v_F \approx 10^6$ m/s is the Fermi velocity and $k$ the wavevector), $G$ was theoretically predicted (S6-S10) to exhibit a universal value $G_0 \equiv e^2/4\hbar$, if the photon energy $E$ is much larger than both temperature and Fermi energy $\varepsilon_F$. Both conditions are stringently satisfied in our visible-optics experiments. The universal value of $G$ also implies that all optical properties of graphene (its transmittance $T$, absorption $P$ and reflection $R$) can be expressed through fundamental constants only ($T$, $P$ and $R$ are unequivocally related to $G$ in the 2D case). In particular, it was noted by Kuzmenko et al (S9) that $T = (1+2\pi G_0/c)^2 = (1+0.5\pi \alpha)^2 \approx 1–\pi \alpha$ for the normal light incidence. We emphasize that – unlike $G$ – both $T$ and $R$ are observable quantities that can be measured directly by using graphene membranes.
To find accurate absolute values of $T$ and $G$, in the analysis shown in Figs. 1B and S2, we have omitted the part of the transmittance spectra at $\lambda < 450$ nm, which as discussed above was affected by hydrocarbon contamination. Also, our apparatus noise was somewhat higher in the infrared region so that, after including the infrared data, the statistical error usually grew rather than decreased. Accordingly, we restricted the analysis to $\lambda < 800$ nm to maximize the accuracy. As a result, we have found $T \approx 97.7\%$ with an accuracy of $\pm 0.1\%$ (see Fig. 1B). The related analysis for $G$ yields $G \approx 1.01G_0$ over the white-light region ($450$ nm $< \lambda < 800$ nm) and the statistical standard error of $\pm 4\%$ (Fig. S2).

The approximation of 2D Dirac fermions is valid only close to the Dirac point and, for higher energies $\varepsilon$, one has to take into account such effects in graphene’s band structure as triangular warping and nonlinearity ($S11$). The triangular warping is significant even for $E << 1$ eV, and there is little left of the linear Dirac spectrum at $\varepsilon$ approaching 5 eV. Therefore, for visible energies of 2 to 3 eV, which are already comparable with the nearest-neighbor hopping energy $t \approx 3$ eV, one may expect the breakdown of the Dirac-fermion approximation used in the calculations of $G_0$. Accordingly, the only earlier theory analysis that did take into account the finite-$E$ corrections was limited to the infrared region ($S9$). For the purpose of our experiments, we have extended the theory to visible frequencies and, also, took into account the next-nearest-neighbor hopping (the latter was found to result only in minute corrections) (see ref. ($S10$) for details). Figure S4 shows the calculated dynamic conductivity $G$ and light transmittance $T$ with the finite-$E$ effects included. One can see a noticeable increase in $G$ at finite $E$ with respect to its idealized value of $\varepsilon^2/4\hbar$ but the corrections still do not exceed 2% for green light. Note that, in the infrared region, the corrections do not disappear but decrease relatively slowly (as $\propto E^2$), until one needs to take into account finite temperature and $\omega_F$ ($S6$-$S10$). Our calculations are also in quantitative agreement with the earlier analysis for $E < 1$ eV ($S9$).

Now we turn our attention to few-layer graphene. It is surprising that its opacity is proportional to the number $N$ of layers involved, at least, to a good approximation for $N \leq 4$ (see the inset in Fig. 1B). Indeed, electronic structures of the multilayer materials are different for different $N$. Generally, several energy bands are present for $N \geq 2$, and the interband distance is given by the energy of inter-plane hopping, $t_\perp \approx 0.3$ eV. This leads to complicated optical spectra with marked absorption peaks corresponding to interband transitions ($S9$, $S12$). However, for visible photon energies $E >> t_\perp$, the spectra significantly simplify so that up to corrections of the order of $(t_\perp/E)^2 << 1$ multilayer graphene can be considered as a stack of independent graphene planes. This leads to the opacity $(1 - T) \approx N\pi\alpha$. This expression was explicitly derived for bilayer graphene $N = 2$ ($S10$) and, also, is apparent from Fig. 1 of ref. ($S12$). Further theoretical analysis is required for few-layer graphene.

**Absorption of light by 2D Dirac fermions**

Finally, we show how the universal value of graphene’s opacity can be understood qualitatively, without calculating its dynamic conductivity first. Let a light wave with electric field $\mathbf{\Theta}$ and frequency $\omega$ fall
perpendicular to a graphene sheet of a unit area. The incident energy flux $W_i$ is given by $W_i = \frac{c}{4\pi} |\Theta|^2$. Taking into account the momentum conservation $k$ for the initial $|i\rangle$ and final $|f\rangle$ states, only the excitation processes pictured in Figure S5 contribute to the light absorption. The absorbed energy $W_a = \eta \hbar \omega$ is given by the number $\eta$ of such absorption events per unit time and can be calculated by using Fermi’s golden rule as $\eta = \frac{(2\pi/\hbar) |M|^2 D}{\pi}$. Taking into account the valley degeneracy, our calculations yield $|M|^2 = |<f| v_F \vec{\sigma} \cdot (\hat{\vec{p}} - \frac{\epsilon}{c} \hat{\vec{A}}) |i\rangle|^2 = \frac{1}{8} e^2 v_F^2 \frac{|\Theta|^2}{\omega^2}$.

This results in $W_a = \left(e^2/4\hbar\right) |\Theta|^2$ and, consequently, absorption $P = W_a/W_i = \pi e^2/\hbar c = \pi\alpha$, both of which are independent of the material parameter $v_F$ that cancels out in the calculations of $W_a$. Also note that the dynamic conductivity $G \equiv W_a/|\Theta|^2$ is equal to $e^2/4\hbar$. Because graphene practically does not reflect light ($R \ll 1$ as discussed above), its opacity $(1 - T)$ is dominated by the derived expression for $P$.

In the case of a zero-gap semiconductor with a parabolic spectrum (e.g., bilayer graphene at low $\epsilon$), the same analysis based on Fermi’s golden rule yields $P = 2\pi\alpha$. This shows that the fact that the optical properties of graphene are defined by the fundamental constants is related to its 2D nature and zero energy gap and does not directly involve the chiral properties of Dirac fermions.

On a more general note, graphene’s Hamiltonian $\hat{H}$ has the same structure as for relativistic electrons (except for coefficient $v_F$ instead of the speed of light $c$). The interaction of light with relativistic particles is described by a coupling constant, a.k.a. the fine structure constant. The Fermi velocity is only a prefactor for both Hamiltonians $\hat{H}_0$ and $\hat{H}_{\text{int}}$ and, accordingly, one can expect that the coefficient may not change the strength of the interaction, as indeed our calculations show.
SUPPLEMENTARY FIGURES

Figure S1. 50 µm aperture partially covered by graphene and its bilayer. This is the original photograph from Fig. 1A, as seen directly in transmitted white light in an optical microscope. No contrast enhancement or image manipulation has been used.

Figure S2. Transmittance spectrum of graphene over a range of photon energies $E$ from near-infrared to violet. The blue open circles show the data obtained using the standard spectroscopy for a uniform membrane that completely covered a 30 µm aperture. For comparison, we show the spectrum measured using an optical microscope (red squares). The red line indicates the opacity of $\pi \alpha$. Inset: Dynamic conductivity $G$ of graphene for visible wavelengths (symbols) recalculated from the measured $T$. The green curves in both main figure and inset show the expected theoretical dependences, in which $G$ varies between 1.01 and 1.04 of $G_0 = \frac{e^2}{4\hbar}$ for this frequency range. The red line and the gray area indicate the statistical average for our measurements and their standard error, respectively: $G/G_0 = 1.01 \pm 0.04$. 

Figure S3. Transmittance spectra of single and bilayer regions of the sample shown in Fig. S1. The transmittance was measured by analyzing images taken in an optical microscope when the membrane was back-illuminated through narrow-band filters.

Figure S4. Dynamic conductivity as a function of photon energy $E$ for graphene, taking into account its triangular warping and nonlinearity at finite energies $\varepsilon$. The curves are given for 3 values of $t$ which cover the possible range expected for this hopping parameter. The corresponding curves for light transmittance are also shown. The red dashed line indicates the value for the idealized case of 2D Dirac fermions.
Figure S5. Excitation processes responsible for absorption of light in graphene. Electrons from the valence band (blue) are excited into empty states in the conduction band (red) with conserving their momentum and gaining the energy $E = \hbar \omega$.

SUPPLEMENTARY REFERENCES