

Supplementary Information *for* Anomalous optical response of graphene on hexagonal boron nitride substrates

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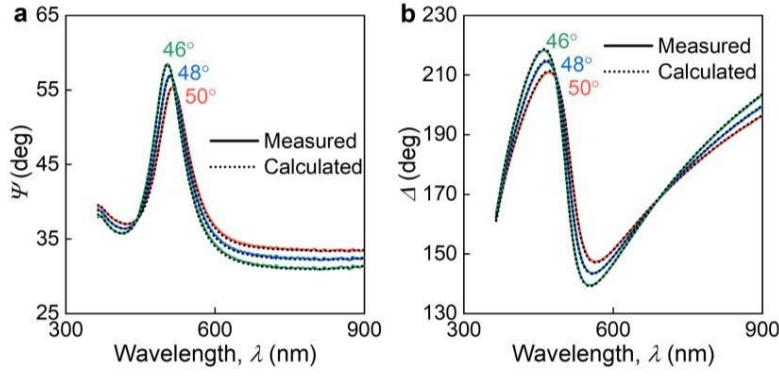
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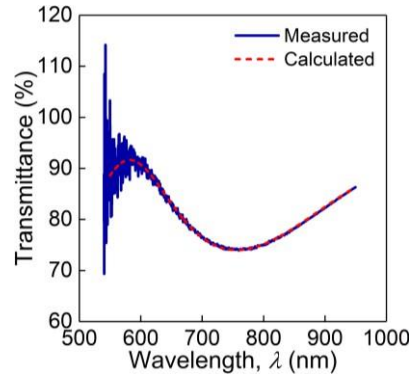
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Supplementary Figure 1: Ellipsometric parameters Ψ (a) and Δ (b) of monolayer graphene on SiO_2/Si substrate. Measurements made at three incident angles of 46° , 48° , 50° . Solid (dashed) lines represent the measured (calculated) cases.

Supplementary Table 1. Drude-Lorentz parameters of graphene samples.

Sample	$\varepsilon_{1\infty}$	A	E_0 (eV)	B (eV)	ρ ($10^{-4} \Omega \cdot \text{cm}$)	r (10^{-3} sec)
Gr/hBN (Sample 1)	0.126	16.7	4.54	0.963	3.26	3.67
Gr/ SiO_2/Si (Sample 2)	1.354	6.29	4.64	1.330	5.50	0.22
Gr/hBN (Sample 3)	0.972	16.6	4.54	0.972	3.21	0.77



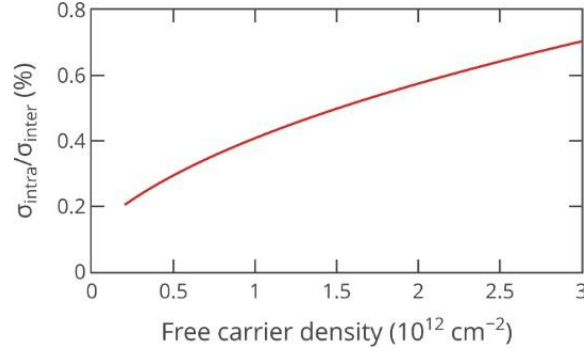
Supplementary Figure 2: Micro-transmittance spectra of hBN on glass substrate. Solid (dashed) line represents the measured (calculated) case.

Supplementary Note 1: Graphene doping vs its absorption.

To estimate the possible contribution from free charge carriers, we have determined the intraband optical conductivity as a function of electron density using similar approach to one presented in¹:

$$\sigma_{\text{intra}} = \frac{2ie^2k_B T}{\pi\hbar^2(\omega + i\tau^{-1})} \ln \left[2\cosh\left(\frac{\mu}{k_B T}\right) \right], \quad (1)$$

where e , $\hbar$ are the elementary charge and the reduced Planck's constant, $k_B T$ is the thermal energy, ω is the angular frequency of light, τ is the electron scattering time, μ is the chemical potential related to the electron density n as $\mu \approx \sqrt{\pi n} \hbar v_F$, where the Fermi velocity is $v_F = 10^6$ m/s. The calculated ratio of intraband to interband optical conductivity, depicted in Supplementary Figure 3, shows that the ratio stays well below 1 % even at an enormous densities of free charge carriers of $3 \times 10^{12} \text{ cm}^{-2}$, which allows us to neglect the possibility of doping of graphene in our work.



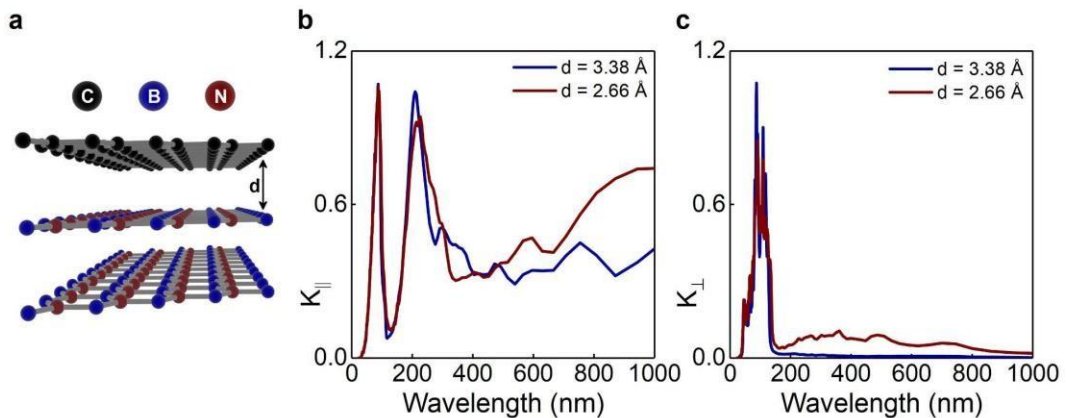
Supplementary Figure 3: Ratio of intraband to interband optical conductivity. Calculations made at $\lambda = 800$ nm as a function of the free charge carrier density calculated assuming $\mu \gg k_B T$.

Supplementary Note 2: *ab-initio* optical response of graphene/hBN heterostructures.

We performed a set of *ab-initio* calculations to study the effects of hBN influence on optical response of monolayer graphene varying their interlayer distances from 3.38 Å (relaxed geometry) down to 2.66 Å with 0.24 Å step. For each of the boundary cases the extinction coefficients are shown in Supplementary Figure 4. Notably, at 2.66 Å, those show animal behaviour in transverse direction at the wavelength range from 200 to 800 nm. Here, we employed a simple model of layered structure and ignored in-plane lattice mismatch between graphene and hBN in-plane parameters. For calculations, we considered 2×2 supercell of graphene and hBN to decrease mechanical stresses during geometry minimization. The total lattice mismatch did not exceed 0.8 % for both graphene and hBN. Geometry optimization of graphene/hBN heterostructures were carried out through density functional theory (DFT)^{2,3} within the generalized gradient approximation (GGA) with Perdew -Burke-Ernzerhof (PBE) parameterization for exchange-correlation functional⁴ implemented in VASPpackage⁵⁻⁷.

Van der Waals interactions were taken into account through Grimme correction⁸. The cutoff energy of plane waves was set to 520 eV. The sampling of the Brillouin zone was done with a uniform k -points grid of 6×6×1. The extinction coefficient was calculated using the superposition of Lorentz oscillators to simulate the complex frequency-dependent dielectric function. The real part was determined *via* the Kramers-Kronig relations, while the imaginary part was found by summing over unfilled states⁹. The extinction coefficient was expressed through the complex dielectric function $\epsilon_1 + i\epsilon_2$ as

$$K(\lambda) = \sqrt{(\sqrt{\epsilon_1^2 - \epsilon_2^2} - \epsilon_1)/2} \quad (2)$$



Supplementary Figure 4: Optical response of graphene/hBN heterostructure vs distance between comprising layers. (a) Schematic illustration of monolayer graphene on top of bilayer hBN used for *ab-initio* evaluations. Evaluated extinction coefficient for (b) *ab*-plane and (c) *c*-axis. Calculations were carried out for different distances between the graphene and hBN, at $d = 2.66$ Å (red line) and $d = 3.38$ Å (blue line), which corresponds to the relaxed geometry.

Supplementary References

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