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# Solving the Controversy on the Wetting Transparency of Graphene

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**Since its discovery, the wetting transparency of graphene, the transmission of the substrate wetting property over graphene coating, has gained significant attention due to its versatility for potential applications. Yet, there have been debates on the interpretation and validity of the wetting transparency. Here, we present a theory taking two previously disregarded factors into account and elucidate the origin of the partial wetting transparency. We show that the liquid bulk modulus is crucial to accurately calculate the van der Waals interactions between the liquid and the surface, and that various wetting states on rough surfaces must be considered to understand a wide range of contact angle measurements that cannot be fitted with a theory considering the flat surface. In addition, we reveal that the wetting characteristic of the substrate almost vanishes when covered by any coating as thick as graphene double layers. Our findings reveal a more complete picture of the wetting transparency of graphene as well as other atomically thin coatings, and can be applied to study various surface engineering problems requiring wettability-tuning.**

Graphene has been a subject of intense research on the basis of its superior optical transparency, electrical conductivity, and mechanical strength[1–6]. Recently, Rafiee *et al.* reported another superior property of graphene, "wetting transparency", which implies that the van der Waals (vdW) interaction between graphene and any liquid placed on top of it is negligible, allowing the "transmission" of the substrate contact angle above graphene. The graphene wetting transparency was spotlighted because of its versatile potential applications[7–10]. Yet, Shih *et al.*[11] showed that the contact angle is significantly affected by the vdW interaction of monolayer graphene and the wetting of the substrate is partially transmitted only for substrates with moderate contact angles (40° ~ 90°). With addition of more graphene layers, the wetting transparency diminishes as the vdW interaction by graphene becomes dominant over the vdW interaction by the substrate.

Although Shih *et al.*[11] established a framework to understand and analyze the wetting transparency, the theory can be improved by incorporating two critical factors that are very important in modeling real experiments. First, the Boltzmann distribution was used to model the water density profile without considering the bulk modulus, a measure of resistance to hydrostatic compression, of the liquid. If a few mathematical flaws are corrected, the theory predicts unrealistically high water density (up to an order of magnitude larger than the ambient density) above the graphene-covered hydrophilic surface. Second, while the theory considered a completely flat surface, it can be improved to explain a wide range of experimental data for surfaces with unknown roughness, if various wetting modes are concerned.

In this work, we report a unifying framework to account for the bulk modulus of the liquid as well as the surface roughness to realistically describe the wetting phenomena. First, we correct a few mathematical mistakes of the previous work in calculating the van der Waals (vdW) interaction energy between the liquid and the substrate. With the corrections, the previous theory predicts zero contact angle for any

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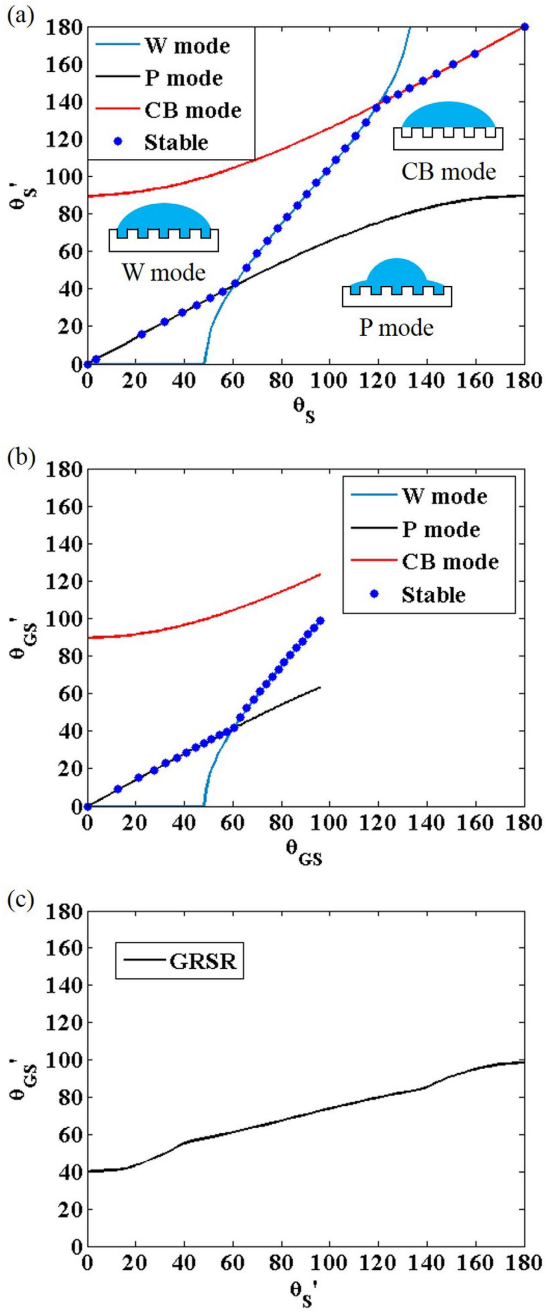


**Figure 2. Illustration of the contact angle calculation for graphene-covered rough surface for the GRGR model with r = 1.5 and $f = 0.5$.** (**a**) Conventional possible wetting modes of the bare substrate, and the actual path of which possesses the minimum free energy[14] (**b**) Conventional possible wetting modes of the graphene covered substrate and the actual path. Note that $\theta'_{GS}$ cannot exceed the contact angle of graphene (96°) (**c**) Contact angle change due to the existence of a graphene film of GRSR model.

| | Bare substrate | GRSR | GFSR |
|---|---|---|---|
| Penetrate mode | $\cos\theta'_S = f\ \cos\theta_S - f + 1$ | $\cos\theta'_{GS} = f\ \cos\theta_{GS} - f + 1$ | — |
| Wenzel mode | $\cos\theta'_S = \mathrm{r}\cos\ \theta_S$ | $\cos\theta'_{GS} = \mathrm{r}\ \cos\theta_{GS}$ | — |
| Cassie Baxter mode | $\cos\theta'_S = f\cos\theta_S + f - 1$ | $\cos\theta'_{GS} = f\ \cos\theta_{GS} + f - 1$ | $\cos\theta'_{GS} = f\ \cos\theta_{GS} + \ (1-f)\cos\theta_G$ |

**Table 1. Contact angles of three roughness models of graphene covered solids (Bare substrate, GRSR, GFSR) due to the wetting modes.**

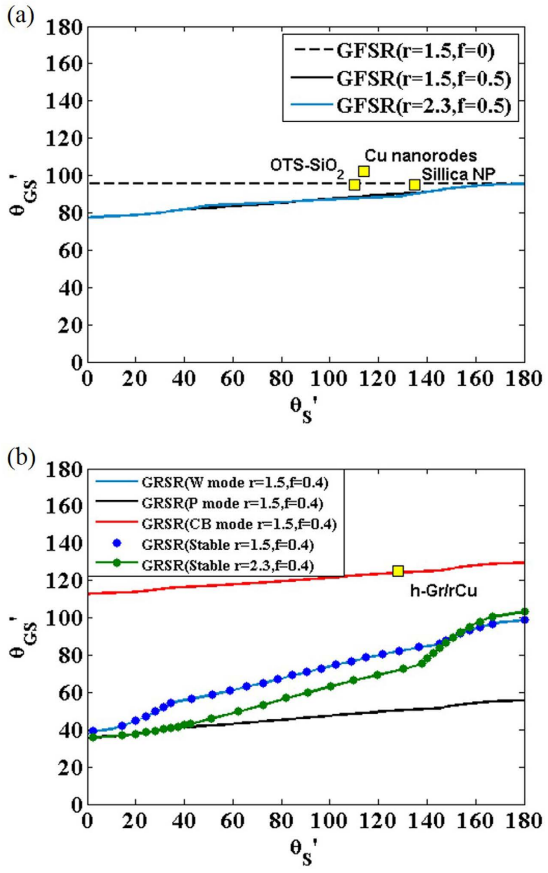


**Figure 3. Theoretical predictions of contact angles of graphene on a rough substrate.** (**a**) Predicted $\boldsymbol{\theta'_{GS}}$ as a function of $\boldsymbol{\theta'_{S}}$ for GFSR model[11,13]. The measurements lie within the curves with $f \leq 0.5$ regardless of the choice of r. (**b**) Predicted $\boldsymbol{\theta'_{GS}}$ for possible wetting modes as a function of $\boldsymbol{\theta'_{S}}$ for GRSR model[12]. The data cannot be explained by the equilibrium curve because the upper bound of $\boldsymbol{\theta'_{GS}}$ is **103.5°** within the reasonable r range of $\mathbf{1 \leq r \leq 2.3}$[23].

the value of $\rho_S A_{SL}$ from 0 to $11.7\,eV\text{Å}^3$, and compute $\boldsymbol{\theta_{GS}}$ for the corresponding range of $\rho_S A_{SL}$ to get $\boldsymbol{\theta_{GS} - \theta_S}$ relationship. Then, the equilibrium wetting mode on the rough surface is determined by the minimum free energy principle, as demonstrated for the GRSR model with r = 1.5 and f = 0.5 in Fig. 2. On the hydrophilic surface ($\theta < 90°$), the equilibrium angle is given by the higher contact angle between the predictions by P mode and W mode. In contrast, the equilibrium angle on the hydrophobic surface ($\theta > 90°$) is chosen as the lower angle between the predictions by W mode and CB mode[14]. According to this principle, we calculate the equilibrium angles $\boldsymbol{\theta'_{S}}$ on the rough bare substrate (Fig. 2a) and $\boldsymbol{\theta'_{GS}}$ on the rough graphene-covered substrate (Fig. 2b). Combining the two predictions, we can plot the relationship between $\boldsymbol{\theta'_{S}}$ and $\boldsymbol{\theta'_{GS}}$ (i.e. pairs of $\boldsymbol{\theta'_{S} - \theta'_{GS}}$ that correspond to identical $\rho_S A_{SL}$ values), as depicted in Fig. 2c.

To validate the theory for a rough substrate, we compare the theoretical predictions with experimental results[7,11–13]. Since the experiments were conducted on surfaces with unknown roughness, we freely choose r within the range of a few available direct measurements by white light profilometry and scanning electron microscope[23] (1 for flat surface $\leq r \leq 2.3$ for very rough surface), and f within the definition (fraction of bare substrate area wet by the liquid, i.e. $0 < f \leq 1$). It is known that a graphene layer floats on the surface when graphene grown by another surface is transferred[13]. Therefore, we applied the GFSR model for most of the data points including Silica NP, Cu nanorodes, and OTS-SiO2, as depicted in Fig. 3a. We obtain the relation between $\boldsymbol{\theta'_{S}}$ and $\boldsymbol{\theta'_{GS}}$ by following the procedure used in Fig. 2 except that we only considered CB mode when computing $\boldsymbol{\theta'_{GS}}$. Most rough substrates are found to have a contact angle around the contact angle of monolayer graphene 96°, the upper bound for the GFSR mode. The measurements lie within the curves with $f \leq 0.5$ regardless of the choice of r. The contact angle above 96° can be explained only if we consider a more complex model in which rough graphene is non-conformally adhered to the rough substrate (see Supplementary Information).

On the other hand, if a graphene film is directly grown on a rough substrate, the film does not float on the substrate but rather conformally adheres to the rough substrate[12]. We compare the measured contact angle of the conformal graphene film on a rough copper surface (denoted as h-Gr/rCu) with the GRSR

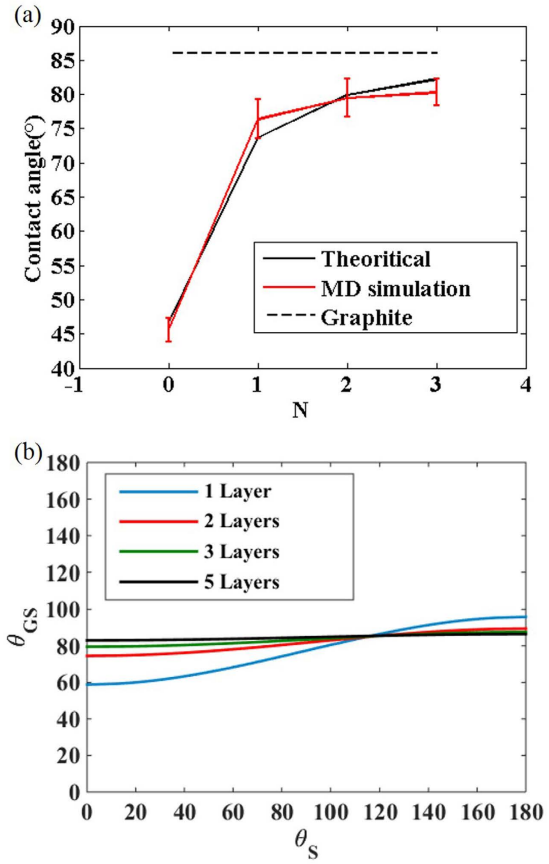


**Figure 4. Theoretical and MD simulation result for wetting of multilayer graphene on a substrate.** (**a**) Predicted $\theta_{GS}$ for composition of N layers of graphene film and solid substrate ($\theta_S = 45°$) (**b**) Predicted $\theta_{GS}$ as a function of $\theta_S$ for composition of 1 ~ 5 layers of graphene film and solid substrate.

model and obtain a very interesting result. While the equilibrium contact mode is predicted to be the W-mode, the predicted equilibrium contact angle curve, $\theta'_S - \theta'_{GS}$ (the blue or green line in Fig. 3b), shows a large discrepancy with the measurement. The data cannot be explained by the equilibrium curve because the upper bound of equilibrium $\theta'_{GS}$ is 103.5° within the reasonable r range of $1 \leq r \leq 2.3$[23]. Interestingly, the experimental result lies on the angle from CB mode with $f = 0.4$. We suspect that the droplet forms the metastable CB mode because of the energy barrier for the transition from the CB state to the air-free W state[14]. Indeed, the original experimental paper[12] also speculated that the high contact angle originated by the CB mode.

**Wetting transparency of multilayer graphene.** We have also studied the wetting transparency of multilayer graphene and find the implication on the wetting transparency of atomically thin coatings. The contact angle of the N-layered graphene covered substrate $\theta_{NGS}$ dramatically converges to the contact angle of graphite 86°[11] as the number of graphene films increases. We reveal that $\theta_{NGS}$ is not substantially affected by $\theta_S$ if more than two layers of graphene films are stacked (Fig. 4a,b). This phenomenon can be analyzed by comparing the distance between the substrate and the water molecules. When monolayer graphene is covered on a substrate, the distance between the substrate and the bottom of the liquid is 6.28 Å, and the substrate contributes a non-negligible portion of the vdW interaction between the entire surface and the liquid. The distance becomes more than 9 Å if two layers of graphene are stacked. Except for with super-hydrophilic solid substrates, double graphene layers will contribute the dominant portion of the vdW potential. If even more graphene layers are stacked, the influence of the solid substrate almost vanishes and the contact angle converges to the contact angle of graphite. Our theory is validated by molecular dynamics simulations[24] (see Methods for details). We consider a model substrate having a contact angle of 45°, and measure the contact angle change when 1–3 graphene layers are covered. The molecular dynamics simulations confirm that the contact angle converges very quickly when three or more layers of graphene are stacked (Fig. 4a).

Our finding implies that most related 2D materials would not show any wetting transparency effect. Due to corrugated or puckered structures, monolayer silicene or phosphorene sheets are significantly thicker than the monolayer graphene[25]. Typical transition metal dichalcogenides materials such as $MoS_2$

is 6.5 Å thick[26], which is comparable to the thickness of graphene double layer. Thus, related 2D materials would show very limited wetting transparency expected for double layer graphene. Any atomically thin coating with the thickness comparable to graphene double layer is enough to almost erase the wetting characteristic of the bare solid.

## Discussion

Based on the theoretical and computational analyses, we provide a comprehensive view on the wetting transparency of graphene. At a first glance, the contact angle measurements on bare and graphene-covered surfaces can be interpreted as an evidence for the wetting transparency for a wide range of contact angles (20° ~ 120°) because they scatter around the perfect wetting transparency line. We show that the observed correlation is a coincidence and cannot be explained by a single universal curve. When a flat graphene layer is covered on a flat surface, the wetting behavior can easily be understood by computing the vdW interaction with the liquid modulus effect. The upper bound of $\boldsymbol{\theta}_{\mathbf{GS}}$ is given by the contact angle of the free standing graphene (96°), and we can conclude that flat graphene monolayer is partially transparent for hydrophilic surfaces. However, in the presence of the surface roughness, three different wetting modes must be considered to determine the equilibrium as well as metastable contact mode to explain the experimental data. When a flat graphene layer is covered on a rough surface, the contact angle of the free standing graphene becomes the upper bound of $\boldsymbol{\theta}_{\mathbf{GS}}$. For a wrinkled graphene (with characteristic roughness scale larger than 10 nm) placed on rough bare surfaces, a contact mode with higher $\boldsymbol{\theta}_{\mathbf{GS}}$ can be formed. In addition, we show that the wetting transparency of graphene double or more layer is negligible, which implies that multilayer graphene covered surface can be treated as a pure graphite surface in terms of its wetting characteristics.

In conclusion, we study the wetting transparency of graphene by accurately calculating the vdW interaction energy for various wetting modes. We reveal that the observed partial wetting transparency cannot be explained without accounting for the bulk modulus and the surface roughness. We also find that this partial wetting transparency almost vanishes for double or more layers of graphene sheets. A natural extension of our work is the research on ionized substrates where coulomb interaction becomes significant, and more complex rough surfaces. We believe that our study can provide a more complete picture of the seemingly partial wetting transparency of monolayer graphene and can be applied to understand wetting phenomena in various circumstances.

## Methods

**Correction of mathematical flaws in the previous work.** First, we briefly review the vdW ineraction calculation presented in Shih *et al.*[11] with the same notations, and then correct a few mathematical flaws. The attractive vdW interaction potential between one carbon atom and one liquid molecule is given by $-\frac{A_{CL}}{r^6}$, where r and $A_{CL}$ are the distance and the vdW parameter between the two entities. Summing up the vdW potential from all pairs of a liquid molecule and carbon atoms, the vdW interaction between one liquid molecule and a flat, infinitely large monolayer graphene sheet is given by $w_{GL}(z) = \frac{-\pi\sigma A_{CL}}{2z^4}$, where $\sigma = 4/(\sqrt{3}\,a^2)$ is the surface density of carbon atoms (with the graphene lattice constant a = 2.49 Å) and z is the shortest distance between the liquid molecule and the graphene plane. The vdW interaction for a flat, infinitely large N-layer graphene sheet adds up to $w_{NL}(z) = \frac{-\pi\sigma A_{CL}}{2}\sum_{i=1}^{N}\frac{1}{[z+(i-1)d_0]^4}$, where $d_0$=3.35 Å is the interlayer distance between the graphitic planes. Ignoring the bulk modulus of the liquid, the density profile of liquids molecule were computed from the Boltzmann distribution, $\rho_L(z) = \rho_{L0}\exp[-w(z)/k_BT]$.

The first mistake in the previous work was made in the calculation of the total vdW interaction potential per unit area between the liquid and the contacting N-layer graphene by $\Phi_{NL} = \sum_{i=1}^{N}\left[\int_{\delta_{GL}+(i-1)d_0}^{\infty}\rho_{NL}(z)\,w_{GL}(z)\,dz\right]$. The density profile $\rho_{NL}(z)$ was written as $\rho_{L0}\exp[-w_{NL}(z)/k_BT]$, and $\delta_{GL} + (i-1)d_0$ referred to the shortest distance between the $i^{th}$ layer and the liquid. The shortest distance $\delta_{GL}$ between the uppermost graphene and the liquid was set to be 3.28 Å, according to the MD results. However, the integral in the square brackets underestimated the vdW interaction from all layers except i = 1, because the liquid density at the bottom (i.e. at $z = \delta_{GL} + (i-1)d_0$), was underestimated as $\rho_{L0}\exp[-w_{NL}(\delta_{GL} + (i-1)d_0)/k_BT]$, although it must be $\rho_{L0}\exp[-w_{NL}(\delta_{GL})/k_BT]$, regardless of i. Thus, $\Phi_{NL}$ must be corrected as

$$\Phi_{NL} = \sum_{i=1}^{N}\left[\int_{\delta_{GL}+(i-1)d_0}^{\infty}\rho_{NL}(z-(i-1)d_0)\,w_{GL}(z)\,dz\right] \tag{2}$$

where $\rho_{NL}(z-(i-1)d_0) = \rho_{L0}\exp[-w_{NL}(z-(i-1)d_0)/k_BT]$.

For a similar reason (see Supplementary Information), the total vdW interaction per unit area $\Phi_{SNL}$ between the liquid and a sheet of N-layer graphene supported by a solid substrate was incorrectly computed and must be fixed as follows:

$$\Phi_{SNL} = \sum_{i=1}^{N}\left[\int_{\delta_{GL}+(i-1)d_0}^{\infty} \rho_{SNL}(z-(i-1)d_0)\, w_{GL}(z)\, dz\right] + \int_{\delta_{SNL}}^{\infty} \rho_{SNL}(z-\delta_{SNL}+\delta_{GL})\, w_{SL}(z)\, dz. \quad (3)$$

Here, $\delta_{SNL} = \delta_{GL} + (N-1)d_0 + \delta_{GS}$ is the shortest distance between the liquid and the solid substrate, where $\delta_{GS}$ is the equilibrium contact separation between graphene and the solid. $\rho_{SNL}(z)$ refers to the liquid density profile above the entire surface, and $w_{SL}(z) = -\frac{\pi\rho_S A_{SL}}{6z^3}$ to the vdW interaction between a liquid molecule and the solid substrate. $\rho_S$ is the density of the substrate and $A_{SL}$ is the vdW interaction parameter between one solid atom and one liquid molecule. On the other hand, the vdW interaction per unit area between the liquid and the bare solid was calculated without any mathematical error by $\Phi_{SL} = \int_{\delta_{SL}}^{\infty} \rho_{SL}(z)\, w_{SL}(z)\, dz$, where the liquid density profile is defined by $\rho_{SL} = \rho_{L0}\exp\left[-\frac{w_{SL}(z)}{k_B T}\right]$ and $\delta_{SL}$ is the distance between the liquid and the bare solid. $\boldsymbol{\theta_S}$ is tuned from 0° to 180° by gradually increasing the value of $\rho_S A_{SL}$ from 0 to 11.7 $eV$Å$^3$, and $\boldsymbol{\theta_{GS}}$ is also computed for the corresponding range of $\rho_S A_{SL}$.

**Molecular dynamics simulations.** We stack graphene films on an arbitrary solid substrate with the contact angle 45°, and calculate contact angles as we add more graphene layers on top (Fig. S1a). We prepare 16,000 liquid water molecules and use half-cylindrical shaped liquid droplet to remove the size effect from triple junction[7]. The simulations are carried out in 300K NVT ensemble using LAMMPS package with a time step of 1 fs. The liquid water molecules are modeled by the extended simple point charge model and the bond lengths are constrained by SETTLE algorithm[11]. On the other hand, substrate is modeled by uncharged vdW particles in face-centered-cubic crystal. For simplicity, atoms in graphene layers and substrate are fixed during simulation. We use 1.5 nm cutoff distance when computing the vdW interactions, and pppm method is used for calculating the coulomb interactions. We equilibrate the molecules for 1 ns and collect the contact angle from the boundary of the liquid droplet obtained from the local water density during the following 1 ns (Fig. S1b)[24].

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## Author Contributions

S.R. and N.M.P. designed the research. D.K. and S.R. developed the theoretical models, carried out numerical calculations, and analyzed the results. D.K. performed the molecular dynamics simulations and conducted the simulation data analysis. N.M.P. and M.J.B. contributed the theoretical analysis and predictions, and analyzed the results. D.K. and S.R. drafted the manuscript and all authors contributed to the writing of the manuscript.

## Additional Information

**Supplementary information** accompanies this paper at http://www.nature.com/srep

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