

## The Surface Property of Graphene

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Since its first discovery in 2004 [1], graphene has attracted great interest due to its extraordinary properties [2-4], such as huge surface area, excellent mechanical strength, high flexibility, superb thermal and electrical conductivity, and exceptional room temperature Hall effect. As a star material for more than one decade, all these properties have been comprehensively studied and tens of thousands of literatures have been published correspondingly. However, there is one fundamental area which is surprisingly not well explored, i.e., the surface property of graphene.

Graphene is a two dimensional material with all the atoms on the surface. It is well accepted that the van der Waals force dominates the interaction between graphene and molecule on top of it. Plenty of publications indicate that molecules can self-assemble themselves on top of graphene which has also been assigned as a pristine property of graphene [5,6]. Nevertheless, the most common molecule, water, indicates that the story is not ended here, because of a simple but difficult question: what is the static water contact angle of graphene?

Almost since its discovery, graphene has been regarded as a hydrophobic surface where the static water contact angle was characterized to be around 90 degree [7,8]. This value was even used as a universal parameter to calibrate some instruments. Considering the huge surface area and corresponding high surface energy, graphene in principle cannot be hydrophilic. This puzzle is finally solved that graphene is indeed hydrophilic at the initial stage after growth and airborne contaminants or water adsorption are responsible for the observed hydrophobicity within one hour [9-11]. These conclusions provide new insight towards the obvious contradictory, but is still unable to answer the above question. Although there are some reports claimed that it is around 45 degree by studying the freshly cleaved highly-ordered

pyrolytic graphite (HOPG) [12], it is still not so convincing, because even though contamination is excluded with exercising care, the studied “graphene” is always on top of other graphene/graphite in the proposed system, which will contribute additional van der Waals force. On the other hand, different static water contact angle values have been reported about graphene on various substrates [9,13].

Because it is practically impossible to fabricate large area, clean and suspended, single layer graphene film as required by water contact angle measurement, a lot of efforts have been devoted into exploring the contribution of the underlying substrate. Two main conclusions, transparency [14] (the wettability of graphene is totally determined by the underlying substrate) and translucency [15] (the wettability of graphene is determined by the more hydrophilic one between graphene and the substrate) were drawn with sufficient experimental and theoretical support. Both discoveries were published on the same high impact journal in neighboring months, indicating the great importance and the huge uncertainty of this topic.

It is logically unbelievable that comprehensive experiment design and careful data collection finally lead to the contradictory conclusions, unless the hypothesis or model was wrong at the first stage or we ignored a dominating/contributing factor. Back to the very beginning, the excellent properties of graphene come from its extraordinary structure. The unique features are the  $\pi$  conjugated system and the Dirac core band structure which indicates that the underlying substrate can not only provide van der Waals contribution but also modify the density of state. Quite interestingly, two independent reports were published again on the same high impact journal in the same months on this topic, but this time they had a similar conclusion that the substrate

modified the hydrophilicity of graphene by doping, which shifted the Fermi level of graphene and changed the corresponding density of state [16,17]. The supplement of the additional factor draws the full picture of the graphene hydrophilicity story, at least at the current stage, although we still cannot answer the aforesaid question. Due to the lack of experimental manner, simulation might provide an alternative way. There are quite a lot of simulations studying the van der Waals interaction between water molecule and graphene [18,19], but the conclusions are also diverse based on the programmed parameters. By adding the doping effect, the orientation of water molecule should be taken into consideration [16]. This might be a good way to adjust the existing model and provide the prediction to our real world.

The understanding of the hydrophilicity of graphene is not simply a fundamental science concern. A lot of applications can also be excited by this discovery and the ability to tune it. So far, it has been confirmed that such knowledge is quite useful for the epitaxy growth of inorganic thin film [20] and the laser-patterned transparent conductive electrode [21]. It also has a great potential in the area of coating materials, biological and chemical sensors, and heat transfer surfaces.

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