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Prof. David Sholl
School of Chemical & Biomolecular Engineering
Georgia Institute of Technology

Zürich, October 19th 2017

Dear Professor Sholl,

We are hereby submitting our revised manuscript entitled, "Doping-Driven Wettability of Two-Dimensional Materials: A Multiscale Theory" by Tian Tian, Shangchao Lin (corresponding author), Siyu Li, Lingling Zhao, Elton J.G. Santos, , and myself (Chih-Jen Shih, corresponding author) for your consideration of publication in *Langmuir* as an article. In our manuscript, we report the first theoretical framework that bridges multiscale physical phenomena on the wettability of doped 2D materials, including: (i) the change of 2D materials surface energy (atomistic scale), (ii) the molecular reorientation of liquid molecules adjacent to the interface (molecular scale), and (iii) the electrical double layer formed in the liquid phase (mesoscopic scale). The theoretical framework presented here allows us to quantify the contact angle change upon doping, suggesting clear strategies towards rational modulation of 2D materials' wetting behavior, as well as control over the molecular packing properties on a 2D material-coated surface. Since tunable properties of 2D-materials-based surfaces mediated by doping are crucial for practical applications, and the search for new ways to effectively control their fundamental properties is an active area of research, we believe that our results will attract the attention of a large number of researchers working in the field.

Our original manuscript was evaluated by three reviewers, who all recognized that there are elements of novelty in this report and of broad interest for the readers of *Langmuir*. Reviewer 2 was very positive about our work, and stated: "This is a very interesting theoretical paper on modeling the change of interfacial tension between a liquid and a sheet of 2D monolayer.". Reviewer 3 was also positive and stated: "This is a timely report as the wettability of 2D materials is sensitive to many factors." Both Reviewers 2 and 3 recommend publication in *Langmuir*. On the other hand, Reviewer 1 raised a few questions regarding some details of our model and suggested further revision.

We have found the reviewer's comments/recommendations to be helpful, and have addressed them to the best of our ability within the scope of our manuscript. This is reflected in both our responses to the reviewers 1 and 3, as well as in the changes that we have implemented in the revised manuscript and Supporting Information.

We hope that the Editor and the reviewers will recognize our effort in addressing the reviewers' comments, and look forward to a positive outcome of our revised manuscript.

Sincerely,

Chih-Jen Shih, ETH Zürich