

Supplementary Information for

Metal/Graphene heterobilayers as hydrogen evolution reaction cathodes:

A first-principles study

Gang Zhou

School of Science, Hubei University of Technology, Wuhan 430068, People's Republic of China
Department of Physics, Tsinghua University, Beijing 100084, People's Republic of China

Corresponding Author

E-mail: 995372896@qq.com

The phonon spectrum was calculated on a simplified model, that is, a Rh nanosheet coat with benzene rings, within the usual harmonic approximation. To balance the calculation accuracy and time consuming, a $p(4 \times 4)$ supercell was adopted for the single-layer Rh sheet and a benzene ring was used to mimic the graphene in the simplified model.

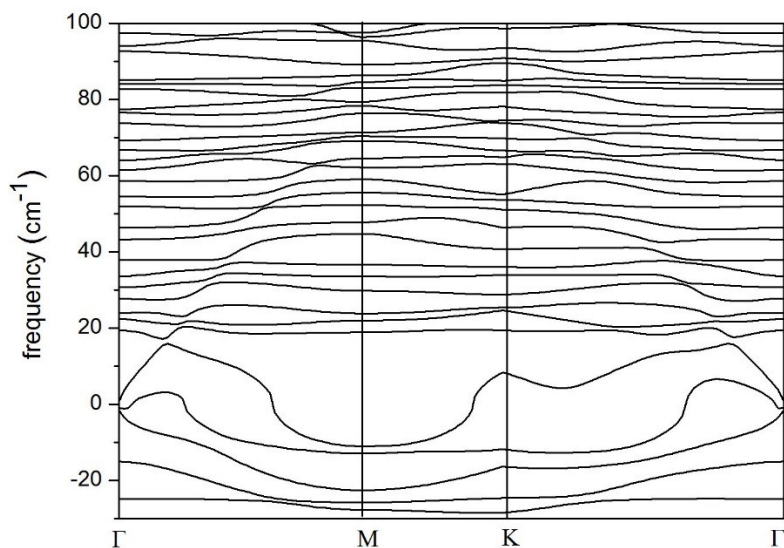


Figure S1 Calculated phonon spectrum of Rh-sheet capped with the benzene rings.

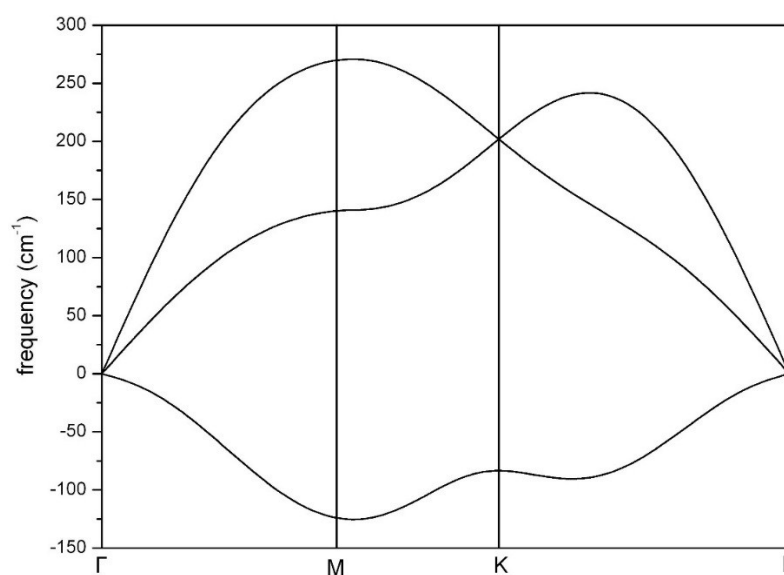


Figure S2 Calculated phonon spectrum of pure single-layer Rh sheet.

By comparing figure S1 with figure S2, we can see that the imaginary frequency modes of single-layer Rh sheet, arising from the out-of-plane motions of all Rh atoms, are largely suppressed by the capped benzene rings. As a result, almost all the imaginary frequencies are significantly reduced to within those of the numerical noise ($< 30 \text{ cm}^{-1}$), indicating that the heterobilayer is thermodynamically stable.