Understanding hydrogen diffusion in between layers of 2D materials

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In 2018, the group of Geim from Manchester performed very interesting experiments, in which hydrogen atoms were transported inside the interstitial space of layered materials, such as hexagonal boron nitride or MoS_2, showing a good sieving of deuterium from protium.[1] We later showed theoretically that indeed hydrogen atoms rather than ions are transported between the layers and reported their diffusion coefficients.[2] We also showed that the transport is assisted by the layer shearing modes. In present work, we investigated the hydrogen diffusion between layers of different transition-metal dichalcogenides (TMDCs), where we studied the influence of possible stackings, stoichiometry, and exemplary twist angles between layers on the self-diffusion coefficients. The calculations were performed using well-tempered metadynamics simulations as implemented in CP2K package,[3] which gives us access to the free energy surface. We found that TMDCs with Se or Mo atoms have lower free energy barriers than these with S or W. Furthermore, structural stackings of MoS_2 (H_h^h(2H), R_h^X (3R), R_h^h, H_h^M, H_h^X) also result in different free energy barriers, varying between 50 and 150 meV, meaning that the corresponding self-diffusion coefficients are in the range of $3.3 - 0.3 \times 10^{-3}$ cm^2 s^{-1}. This study is the first step to understand whether stacking-induced self-diffusion, which most likely depends on the twist angle between layers and the resulting moiré potential, can be optimized to be more directional.

References

[1] S. Hu, K. Gopinadhan, A. Rakowski, M. Neek-Amal, T. Heine, I. Grigorieva, S. Haigh, F. Peeters, A. Geim, and M. Lozada-Hidalgo, Nat. Nanotechnol., 13 (2018) , 468.
[2] Y. An, A. Kuc, P. Petkov, M. Lozada-Hidalgo, and T. Heine, Small 15 (2019), 1901722.
[3] J. Hutter, M. Iannuzzi, F. Schiffmann, and J. VandeVondele, Wiley Interdiscip. Rev. Comput. Mol. Sci. 4 (2014), 15.

Figures

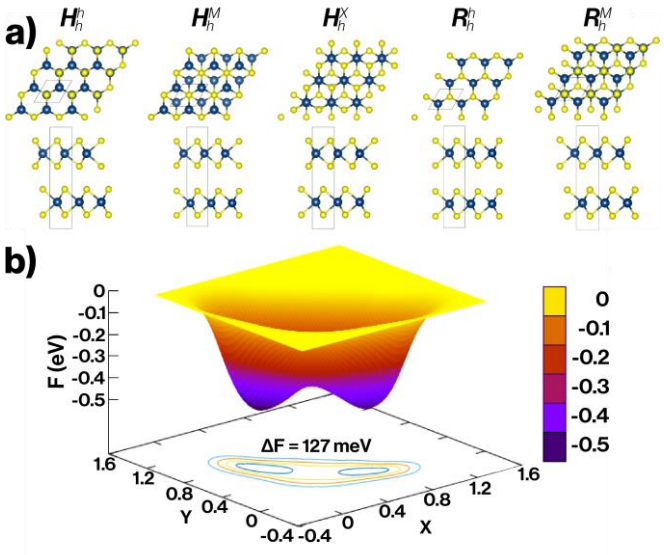

Figure 1: (a) Top and side views of all considered structural high-symmetry stackings. Unit cells are marked with black lines. Blue - Mo, yellow - S.
(b) Free-energy surface (F) of the H transport through interstitial space (also named as collective variables (CV1: X- CV2: y-axis) obtained from well-tempered metadynamics simulations for WS_2.