Constructing Metallic Nanoroad on MoS_2 Monolayer via Hydrogenation

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1. Hydrogen diffusion on graphene

The nudged elastic band (NEB) for hydrogen diffusing on the basal plane of graphene is shown in Figure S1. In the calculation, a 5×5 supercell is used and the method for describing exchange-correlation interaction is generalized gradient approximation (GGA). The obtained barrier is 1.25 eV, consistent with previous report of 1.19 eV.[1] This activation barrier is much larger than the barrier (0.29 eV) for H diffusing on MoS_2. The strong C-H bonding is the underlying reason for the large barrier in graphene whereas the weaker S-H bonding leads to a smaller barrier in MoS_2. It should be noted that the diffusing paths on MoS_2 and graphene are very different although the host lattice of graphene and MoS_2 are both honeycomb lattice. For graphene, the hydrogen atom diffuses between different sublattices along the armchair direction, that is, the A sublattice and the neighboring B sublattice. However, for MoS_2, the hydrogen atom diffuses between the same sulfur sublattice along the zigzag direction.

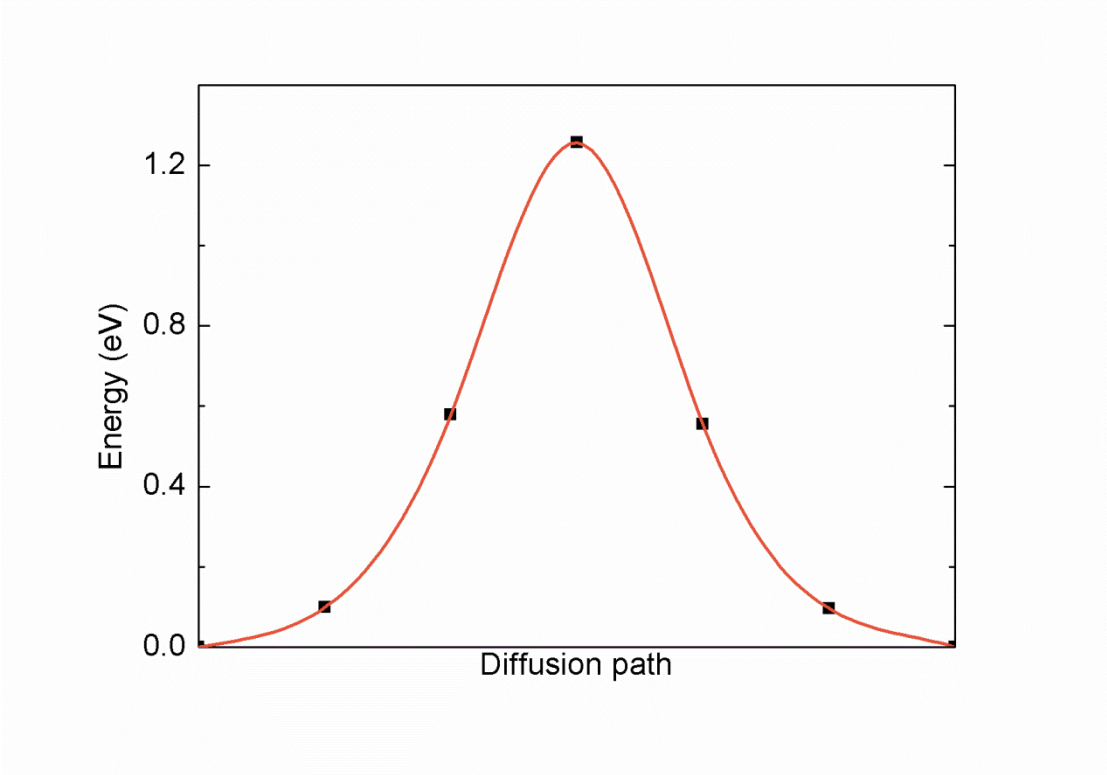

Figure S1. Activation energy of hydrogen atom diffusing on graphene. The NEB diffusion barrier is calculated for hydrogen hopping between two neighboring carbon atoms on graphene sheet.

2. Coupling effect of two H atoms

We test the size effect on the binding energy of the two H atoms on MoS_2. The two configurations with the strongest interaction, that is, 1-CC_{ZZ} and 1-$BA_{ZZ,}$ are calculated using 5×5, 6×6, and 7×7 supercells by the GGA method. The results are listed in Table S1. It can be seen that the difference in energy is less than 0.01 eV.

Table S1. **Binding energy of two H atoms on the MoS_2 monolayer using different supercells.**

Configurations	5×5 (eV)	6×6 (eV)	7×7 (eV)
1-CC_{ZZ}	0.313	0.309	0.306
1-BA_{ZZ}	0.355	0.361	0.368

3. Binding energy of single H on MoS_2

The strength of single H atom binding on the surface is obtained through calculating the binding energy (E^b) of single H atom on MoS_2 monolayer, which is defined as:

$$E^b = E(\mathrm{H{:}MoS_2}) - E(\mathrm{MoS_2}) - E(\mathrm{H}) \quad (1)$$

where $E(\mathrm{H{:}MoS_2})$ and $E(\mathrm{MoS_2})$ are the total energy of H-adsorbed and clean MoS_2, respectively. $E(\mathrm{H})$ is the energy of single H atom. Here, we only consider the most stable configuration for H adsorption with the tilted form in neutral charge state. The calculated E^b are -1.13 and -2.26 eV for GGA and HSE06 methods, respectively, suggesting a strong binding of H on the surface. This result is consistent with previous study.[2]

4. DOS analysis for H vacancies on H:MoS_2 strips

In the case of H adsorption on graphene, the full coverage of H on the surface seems to be impossible. The graphane sheet normally contains H vacancies on the surface [3] and the approach for adjusting the band gap width is currently considered through partial graphene coverage with hydrogen.[4,5] In the case of H:MoS_2 nanoroad, similarly, the most likely defects are the H vacancies on the H strip. To analyze the effect of this type of defect on the metallicity of the nanoroad, we calculate the partial

density of states (PDOS) based on the H:MoS_2 nanoroads along the armchair direction containing 8 H chains. Figure S2(a) is the PDOS for the perfect nanoroad. Figure S2(b) is the PDOS for the nanoroad containing 4 H vacancies on the surface, equivalent to a 50% coverage of H on the nanoroad. It can be seen that the metallic character is still maintained and the metallicity does not degrade with the surface containing 50% H on the nanoroad.

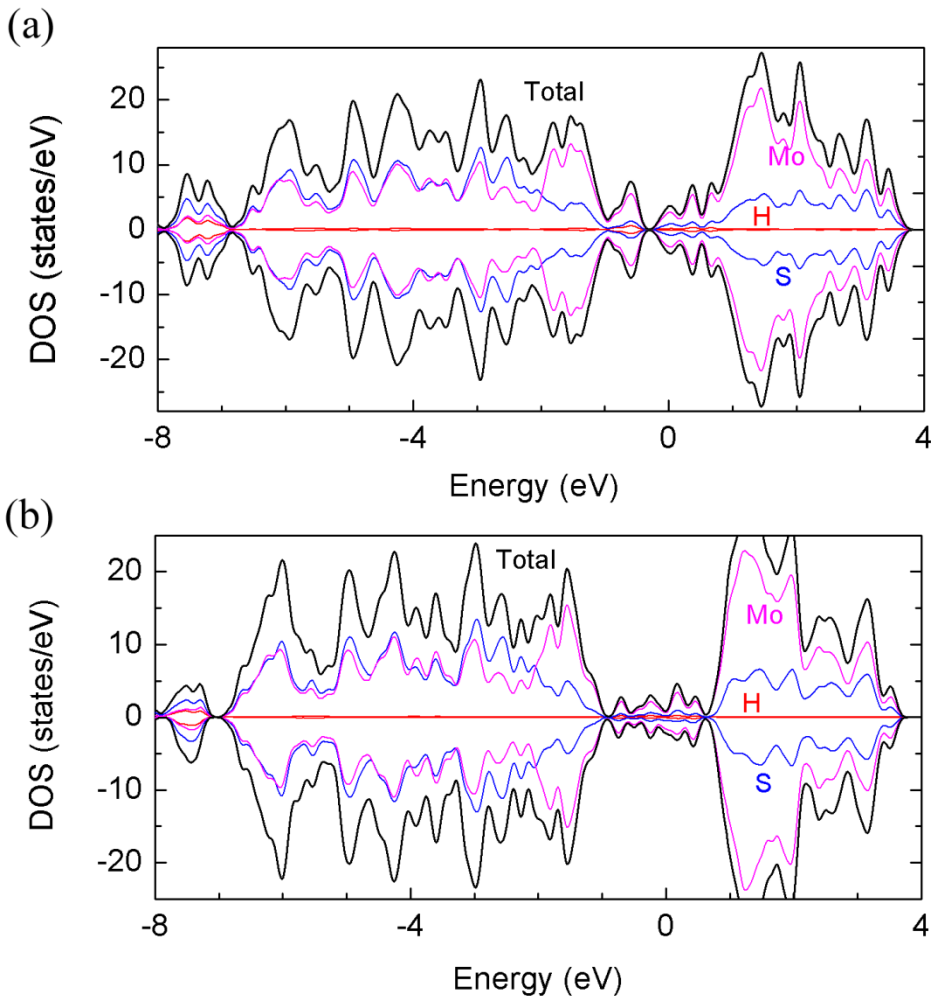

Figure S2. Effects of H vacancies on metallization of H:MoS_2 nanoroad. Spin-polarized calculation based on GGA for (a) PDOS of perfect H:MoS_2 nanoroad along the armchair direction containing 8 H chains, and (b) PDOS of the same

H:MoS_2 nanoroad in (a) but containing 50% H on the nanoroad.

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