Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

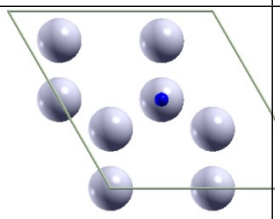
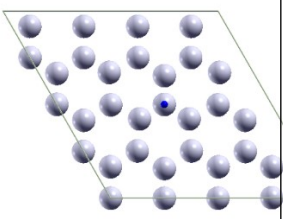
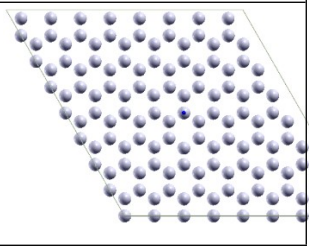
Hydrogen adsorption and diffusion through a two dimen-
sional sheet of Lithium: A first principles study

Shrijan Roy,*[a] Prince Singh,[a‡] Kiran Yadav, [a‡] and Nirat Ray[a]

Department of Materials Science and Engineering, Indian Institute of Technology Delhi, Hauz Khas, New Delhi 110016,
India

Supplementary Information

A. Effect of supercell size on adsorption energies.

Supercell Size	*2*2*1*	*4*4*1*	*8*8*1*
Adsorption Energy at D-site (eV)	*-3.01*	*-3.02*	*-3.05*
			

B. Dispersion effects on the adsorption energies at various sites.

		Without dispersion	*With dispersion effects*
Adsorption Energy (relaxed monolayer) at site (eV)	*D*	*-2.723*	*-3.02*
	H	*-2.707*	*-2.84*
	X	*-2.720*	*-2.95*
	T	*-1.773*	*-1.81*

C. *Adsorption energy, Zero point energy (ZPE) and Gibbs free energy for different sites.*

	D site	*X site*	*H site*	*T site*
E_{ads} *(eV)*	*-3.02*	*-2.95*	*-2.84*	*-1.81*
ZPE (eV)	*0.012*	*0.013*	*0.011*	*0.011*
Gibbs Free energy (eV)	*2.63*	*2.56*	*2.45*	*1.42*

D. *Coverage (upto 1 ML)Figure 2: The large structural distortions in the structure at high coverage for (a) H site and (b) X site.*

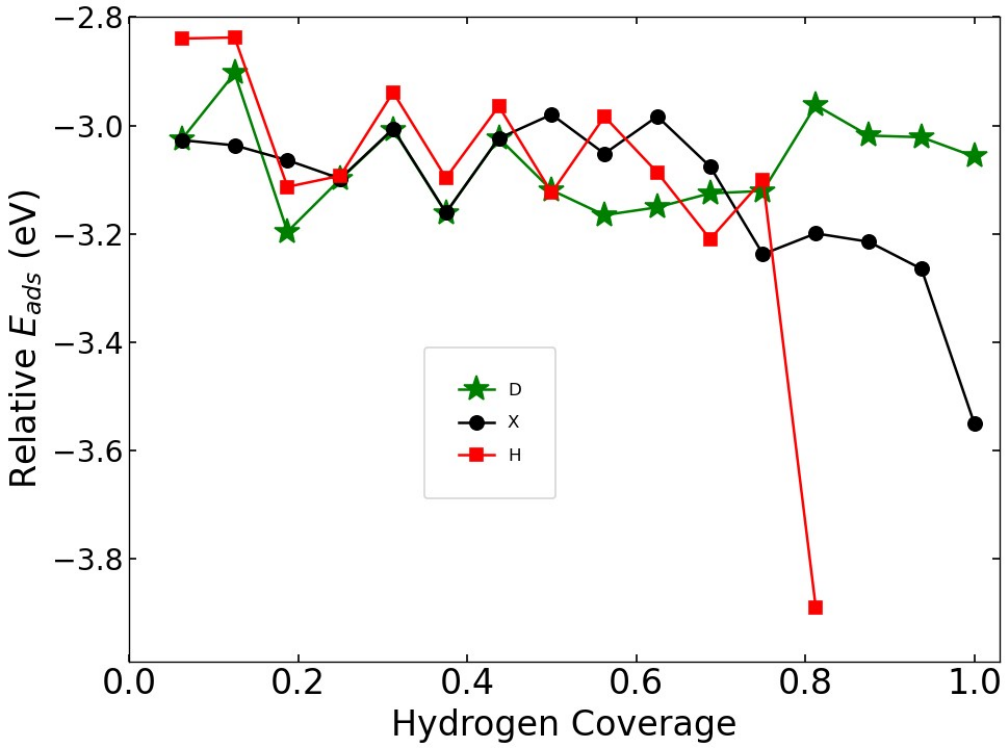


Figure S1: Adsorption energy as a function of coverage. Large structural distortions begin with high coverage for X and H sites, and are correlated with the decline in adsorption energyafter 0.8.

E. NEB calculations for diffusion of hydrogen from D to H site (asymmetric barrier)

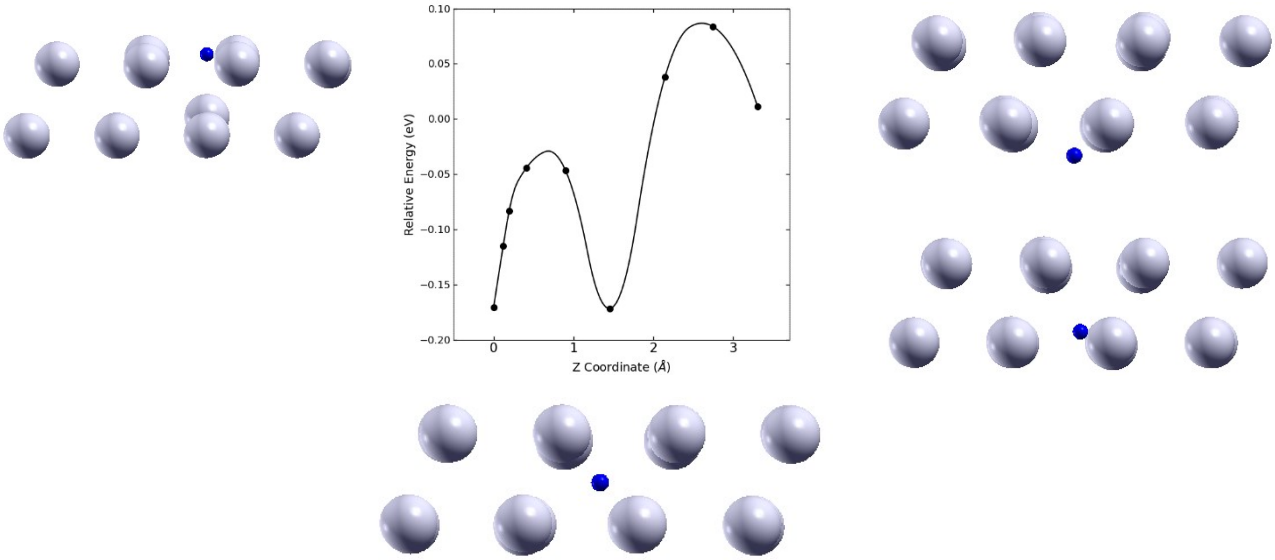


Figure S2: Diffusion of hydrogen atom from D to H.

F. Phonon frequencies for adsorbed hydrogen

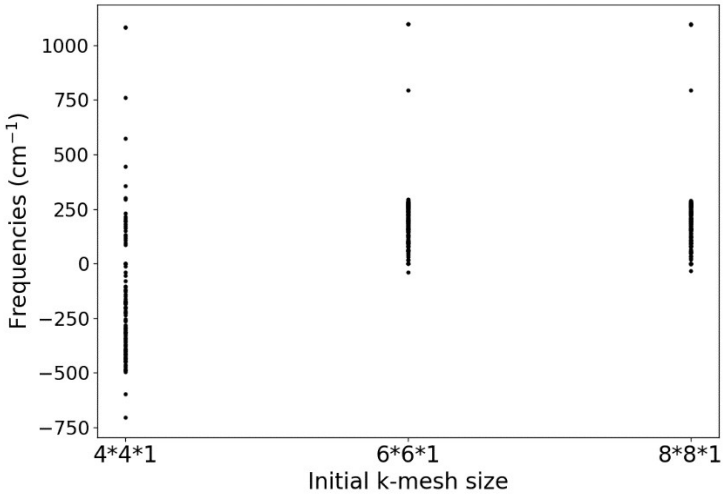


Figure S3: Gamma point phonon frequencies as a function of k-mesh used for calculation.

While the high frequency modes are not impacted by the k-mesh, the low frequency modes are extremely sensitive to the k-mesh. Small instabilities (~1-50 cm-1)are seen even at denser k-mesh values.