Supplementary materials

Diffusivity and band offset analysis of graphene interlayer at back contact of Copper Zinc Tin Sulphide Solar Cell

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Nature of S-C bonds for S adsorbed on pristine graphene:

To understand the nature of the 2 S-C bonds, we computed the charge density of the S atom adsorbed on the graphene bridge site, as shown in Fig. S1. From the charge density, we can see there is no significant overlap of orbitals between the S atom and the two C atoms. Therefore, the S-C bonds are relatively weak with limited charge transfer. We also computed the partial density of state (PDOS) of the S atom and the C atom. The nature of the bonding is metallic, as shown in Fig. S2. From the PDOS, we can see that there is small coupling between S-3p and C-2p near the Fermi level. And there are additional couplings among their s and p orbitals at lower energy states.
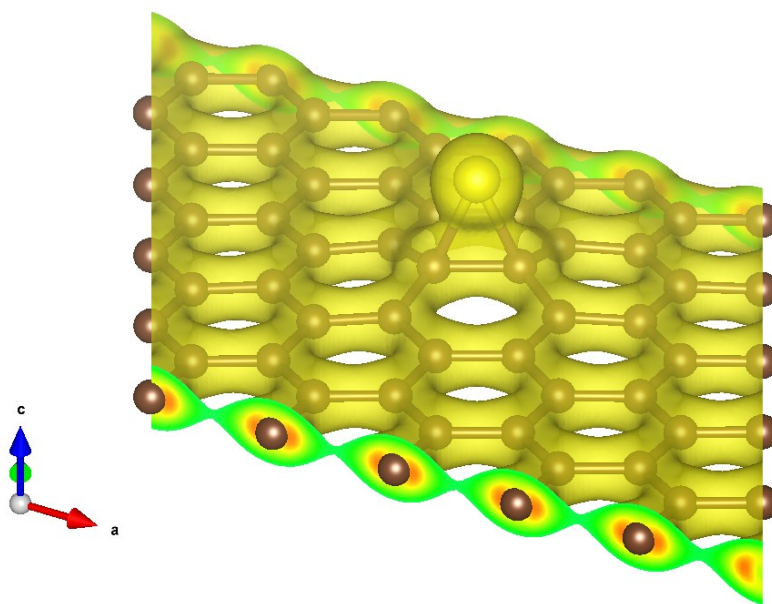

Fig. S1. The charge density of S atom adsorbed on the pristine graphene bridge site.

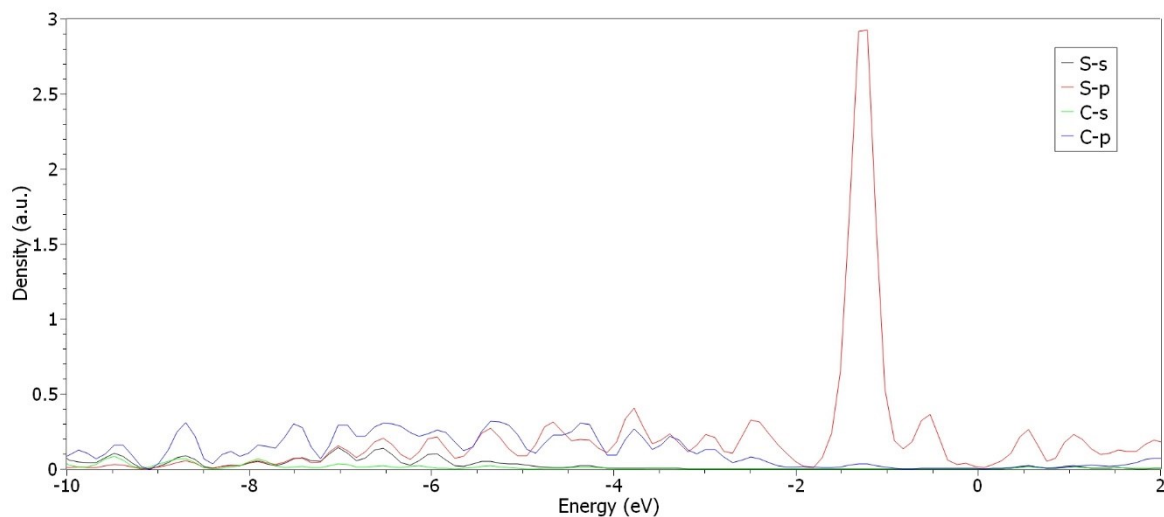[1] Corresponding author, jyzhu@phy.cuhk.edu.hk

Fig. S2. The PDOS of the S atom and the C atom.

Nature of S-C bonds in transition states:

For more than two S-C bonding cases, we choose the saddle point of S atom diffusing across pristine graphene and computed the charge density, as shown in Fig. S3. The bonding nature is multicentered and metallic. We further calculated the PDOS and found that the bonds are due to the overlaps between the hybridized sp orbitals of S and that of C, as shown in Fig. S4.
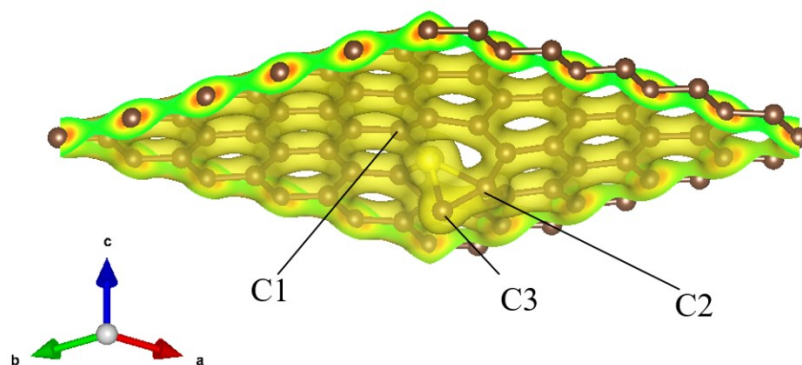

Fig. S3. The Charge density of the saddle point of S atom diffusing across pristine graphene
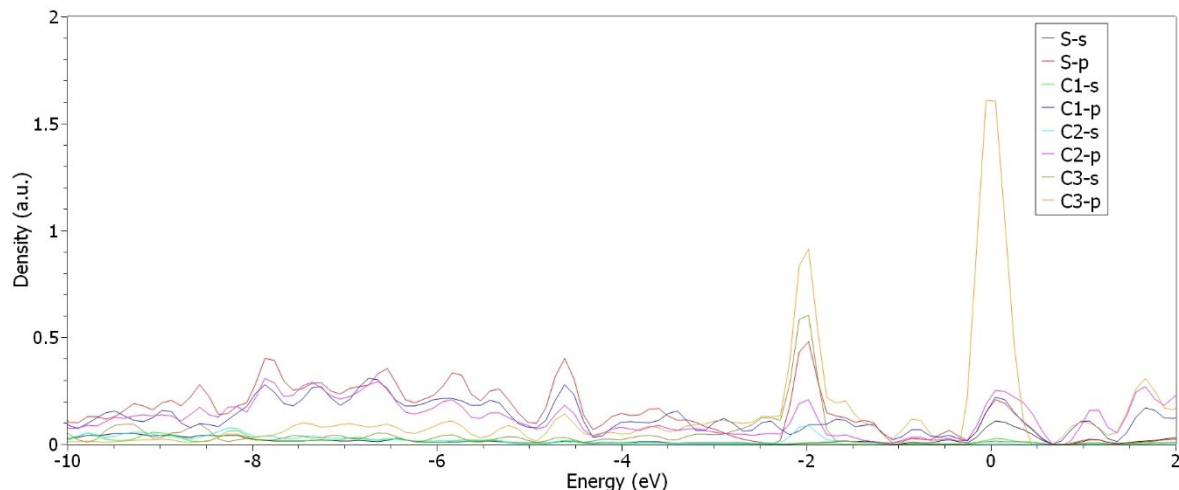
Fig. S4. The PDOS of the 3 labeled C atoms and the S atoms in Fig. S3. There is one extra C atom with similar PDOS to C1 due to symmetry, which is not shown.

Relative formation energies of S on Stone-Wale defect:

Table SI. Relative formation energies of different S atom adsorption sites as shown in Fig. 8.

Adsorption site	Formation energy (eV) relative to the formation of an S atom on the single vacancy
1	5.41
2	5.75
3	5.70
4	6.20
5	6.60
6	6.12

Partial charge densities of pseudohydrogens and VBM:

To show the localization of the surface charge density of the pseudohydrogens, we computed the partial charge density of them with fractional charge of q=1.75e, q=1.5e, and q=1e, as shown in Fig. S5 (a), (b), and (c), respectively. The hydrogen states are localized near the $(11\overline{2})$ surface and does not affect the $(\overline{1}\overline{1}2)$ surface. We also computed the partial charge densities of the VBM as shown in Fig. S5 (d), which are widely distributed in the bulk region.

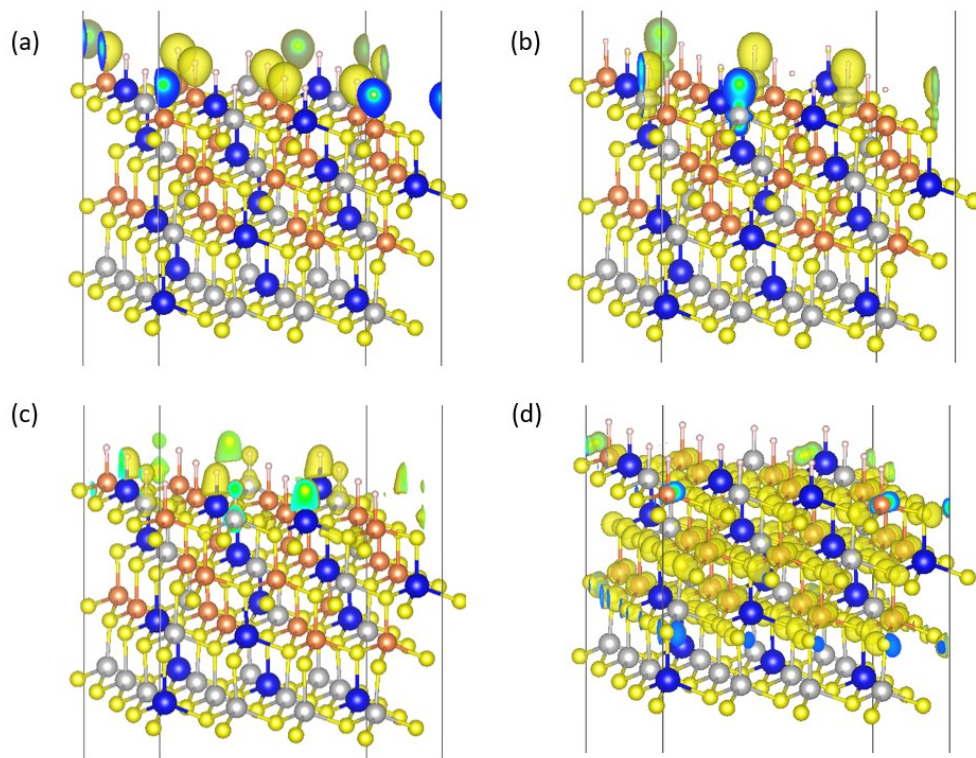Fig. S5. (a) Partial charge density of q=1.75e pseudohydrogen. (b) Partial charge density of q=1.5e pseudohydrogen. (c) Partial charge density of hydrogen. (d) Partial charge density of VBM.