Supporting Information

First-Principles Calculations on Resistance and Electronic Properties of H_2 Adsorption on CoO-SnO$_2$ Heterojunction Surface

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The n-type and p-type semiconductor materials have the different charge carrier, therefore, the space charge models of n-type semiconductor materials and p-type semiconductor materials need to be discussed respectively. The electrons will transfer from the surface of the semiconductor material to oxygen when the metal oxide semiconductor material be exposed in air. Oxygen will capture the electrons and form negative oxygen anion.

Because the concentration of electrons in n-type semiconductor materials is larger than the concentration of holes, n-type semiconductor materials are electrically conductive. When n-type materials are exposed to the air, the oxygen in the air will capture the electrons on the surface of n-type semiconductor materials to form adsorbed oxygen anion. And the electrons on the surface of n-type semiconductor materials are decrease.

When the n-type metal oxide semiconductor materials are in the reducing gases such as H_2, the reducing gases will provide electrons to the n-type metal oxide semiconductor materials. It will increase the electrons in the n-type metal oxide semiconductor materials, and decrease the resistance, changing the conductivity to H_2.

 The concentration of electrons in p-type semiconductors is far less than that of holes, so p-type semiconductors conduct holes and holes are charge carriers. When p-type materials are exposed in the air, the oxygen in the air acquires the electrons on the p-type semiconductor materials surface and forms oxygen negative ions. Meanwhile, the electrons on the surface of p-type semiconductor materials decrease and the holes

concentration increases. When p-type metal oxide semiconductor materials are in reducing gases such as H_2, the reducing gases will provide electrons to p-type metal oxide semiconductor materials. It will increase the electrons and decrease the holes concentration in p-type metal oxide semiconductor materials. As a result, the decreases about the carrier concentration, the conductivity, and the resistance increases, changing the conductivity to H_2. Because the adsorption of oxygen will affect the adsorption about target gas on the surface of gas carrier, we need to find out the optimal adsorption point of oxygen on the surface of gas carrier. The smaller E_{ads} of gas on the heterojunction surface, indicated the gas adsorption is more stable and difficult to be dissociated. The calculation result as follow:

Table S1 The adsorption of O_2 on $SnO_2(110)/CoO(100)$ heterojunction

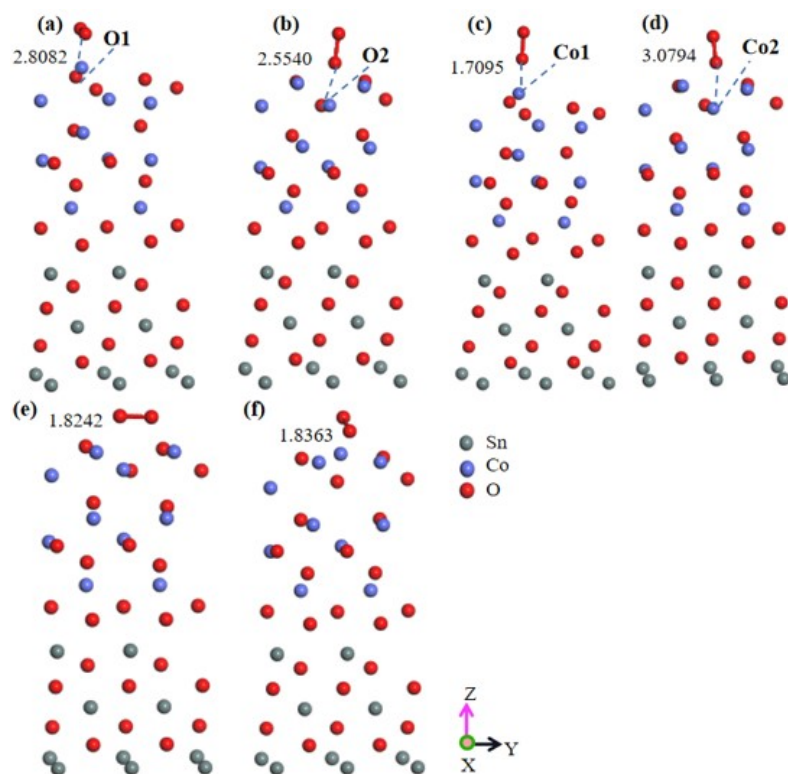| Sites	E_{ads} (eV)	Distance (Å)
O1 vertical	-3.6871	2.8082
O2 vertical	-3.6624	2.5540
Co1 vertical	-3.6624	1.7095
Co2 vertical	-3.5170	3.0794
Parallel surface 1	-4.4046	1.8242
Parallel surface 2	-4.3139	1.8363

Figure S1 Optimized structure that O_2 is adsorbed on $SnO_2(110)/CoO(100)$ heterojunction surface.

(a-d) Vertical the surface; **(e, f)** Parallel surface 1, and Parallel surface 2.

There are vertical and parallel modalities when O_2 is adsorbed on different sites of $SnO_2(110)/CoO(100)$ heterojunction surface (including four vertical formation O1, O2, Co1, Co2 sites and parallel formation Parallel surface 1, Parallel surface 2) as shown in Figure S1. In Table S1, the E_{ads} is negative when O_2 is adsorbed on different sites of the heterojunction surface. It means the adsorption of O_2 on the heterojunction is spontaneous. The adsorbed distance of O_2 on different sites is less than 3 Å, expect on Co2 site. It indicates the formation of bonding between O_2 molecule and adsorbed sites. According to the E_{ads} and adsorbed distance, Parallel surface 1 is the best form of O_2 on the heterojunction surface.

Table S2 The adsorption of O_2 on CoO(110)/SnO$_2$(100) heterojunction

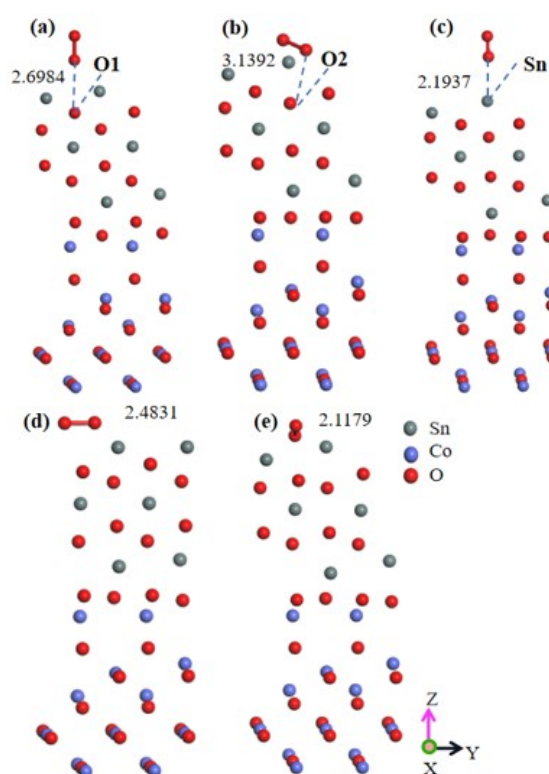| Sites	E_{ads} (eV)	Distance (Å)
O1 vertical	-4.8895	2.7525
O2 vertical	-5.1878	3.0425
Sn vertical	-5.1056	2.7120
Parallel surface 1	-5.4001	2.4031
Parallel surface 2	-5.3621	2.4179

Figure S2 Optimized structure that O_2 is adsorbed on CoO(110)/SnO$_2$(100) heterojunction surface.

(a-c) Vertical the surface; **(d, e)** Parallel surface 1 and Parallel surface 2.

Figure S2 shows vertical and parallel modalities when O_2 is adsorbed on different sites of the CoO(110)/SnO$_2$(100) heterojunction surface (including three vertical formations O1, O2, Sn sites and two parallel formations Parallel surface 1, Parallel surface 2). The E_{ads} is negative when O_2 is adsorbed on different sites of the

heterojunction surface, as shown in Table S2. It indicates the adsorption of O_2 molecule on the heterojunction surface is an exothermic reaction and O_2 can be stably adsorbed on the heterojunction surface. All adsorbed distance are less than 3 Å when O_2 is adsorbed on different sites of the heterojunction surface, except on O_2 molecule, O2 sites. In the case of Parallel surface 1 owes the smaller E_{ads} and less adsorbed distance of O_2 on the heterojunction surface. Consequently, Parallel surface 1 is the best adsorption form of O_2 on the heterojunction surface.

Table S3 H_2 is adsorbed on CoO(110)(II)

Site	E_{ads} (eV)	Distance (Å)
O_2	0.8061	2.5183
O1	0.4413	3.3777
O2	-2.3144	3.8017
Co1	-0.4980	2.5752
Co2	-0.0618	4.3027

Table S4 H_2 is adsorbed on SnO$_2$(100)(II)

Atom	E_{ads} (eV)	Distance (Å)
O_2	-0.2475	2.2459
Sn	-0.2884	2.8692
O1	2.3778	3.5501
O2	-1.0945	2.6711

Table S3 shows the E_{ads} and the adsorbed distance when H_2 is adsorbed on different sites (including O site of O_2 molecule, and O1, O2, Co1 and Co2 sites) of CoO(110) surface. The E_{ads} and adsorbed distance have obvious variations. The adsorption energy is positive when H_2 is adsorbed on O site of O_2 molecule, O2 site, and negative when H_2 is adsorbed on O2 site, Co1 site and Co2 site. It evidently indicates that the adsorption of H_2 on O2 site, Co1 site and Co2 site are exothermic. In addition,

the adsorbed distance is less than 3 Å when H_2 is adsorbed on O_2 molecule and Co1 site. Thus, according to the E_{ads} and adsorbed distance, H_2 tends to be adsorbed on Co1 more easily.

Table S4 reveals the adsorption energy and the adsorbed distance when H_2 is adsorbed on different sites of $SnO_2(100)$ surface, including O site of O_2 molecule, O1 site, O2 site, and Sn (where the surface Sn atoms are equivalent for passing through the supercell) site. The E_{ads} is positive when H_2 is adsorbed on O1 site of $SnO_2(100)$ surface. And the E_{ads} are negative when H_2 is adsorbed on O site of O_2 molecule, O1 site, and O2 site. Moreover, the adsorbed distance are less than 3 Å when H_2 is adsorbed on O site of O_2 molecule, O1 site, and O2 site. Therefore, H_2 not only readily adsorbs to the O2 site of $SnO_2(100)$ surface and O sites of O_2 molecules, but also forms chemical bonds with O2 site and O_2 molecule.

Overall, Table S3 and S4 demonstrate that H_2 readily adsorbs on the SnO_2 surface.

Table S5 The adsorption of O_2 on CoO(110) surface

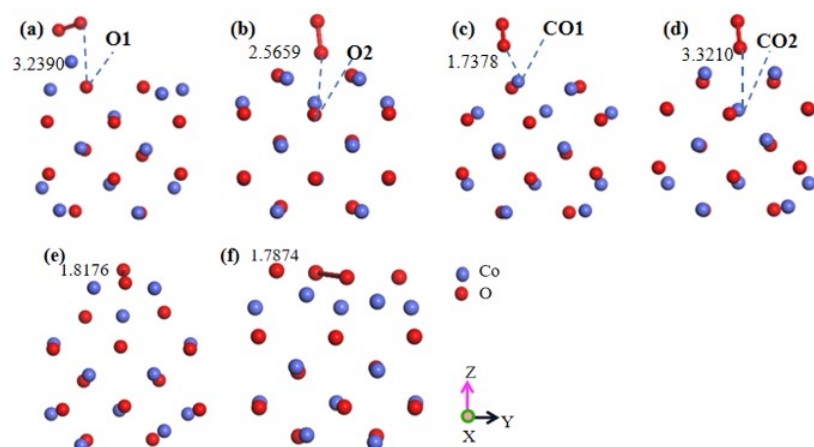| Sites	E_{ads} (eV)	Distance (Å)
O1 vertical	-6.3345	3.2390
O2 vertical	-2.4048	2.5659
Co1 vertical	-2.7532	1.8378
Co2 vertical	-3.2925	3.3210
Parallel surface 1	-4.8428	1.8176
Parallel surface 2	-3.4120	1.7874

Figure S3 O_2 is adsorbed on CoO(110) surface. **(a-d)** Vertical the surface; **(e, f)** Parallel surface 1 and Parallel surface 2.

Figure S3 reflects vertical and parallel modalities when O_2 molecule is adsorbed on CoO(110) surface (including four vertical formation O1, O2, Co1, Co2 sites and two parallel formation Parallel surface 1, Parallel surface 2 respectively). The E_{ads} are negative when O_2 is adsorbed on different sites of CoO(110) surface as shown in Table S5. It reveals the adsorption of O_2 molecule on Co(110) surface is exothermic. The adsorbed distance is more than 3 Å when O_2 is adsorbed O1 site and Co2 site of CoO(110) surface, except O_2 on O2 and Co1 sites. It indicates the formation of bonding between O_2 and different adsorbed sites on O1 sites and Co2 sites. According to the E_{ads} (adsorption energy) and adsorbed distance, Parallel surface 1 is the best adsorption form of O_2 molecule on CoO(110) surface.

Table S6 The adsorption of O_2 on SnO_2(100) surface

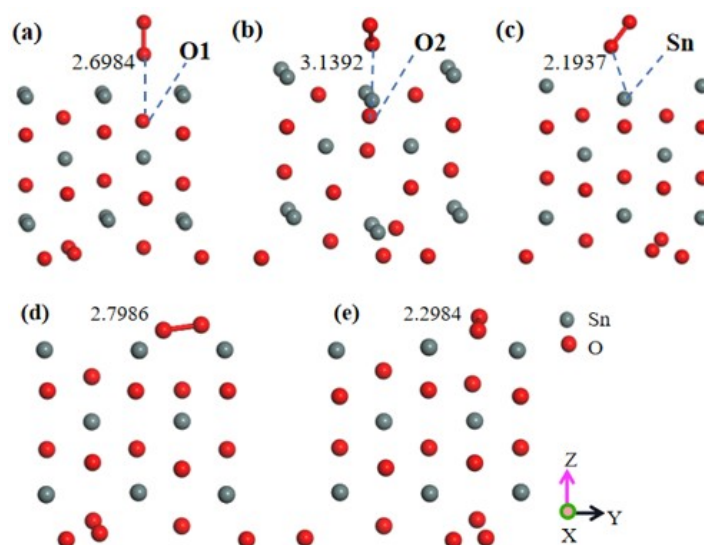| Sites	E_{ads} (eV)	Distance (Å)
O1 vertical	-4.2644	2.6984
O2 vertical	-4.9666	3.1392
Sn vertical	-3.9666	2.1937
Parallel surface 1	-4.5361	2.1986
Parallel surface 2	-4.4308	2.2982

Figure S4 O2 is adsorbed on SnO$_2$(100) surface. **(a-c)** Vertical the surface; **(d, e)** Parallel surface 1 and Parallel surface 2.

Figure S4 shows five patterns when O$_2$ molecule is adsorbed on different sites of SnO$_2$(100) surface (including three vertical formations O1, O2, Sn sits and two parallel formations Parallel 1, Parallel 2). The E$_{ads}$ is negative when O$_2$ is adsorbed on different sites of SnO$_2$(100) surface as shown in Table S6. It indicates that the adsorption of O$_2$ molecule on SnO$_2$ surface is exothermic. The adsorbed distance is less than 3 Å which indicates the formation of bonding between O$_2$ molecule and different adsorbed sites, except on O2 site. According to the E$_{ads}$ and adsorbed distance, in the case of Parallel surface 1, the O$_2$ adsorption is not only stable, but also has the best bonding effect with the adsorption site.
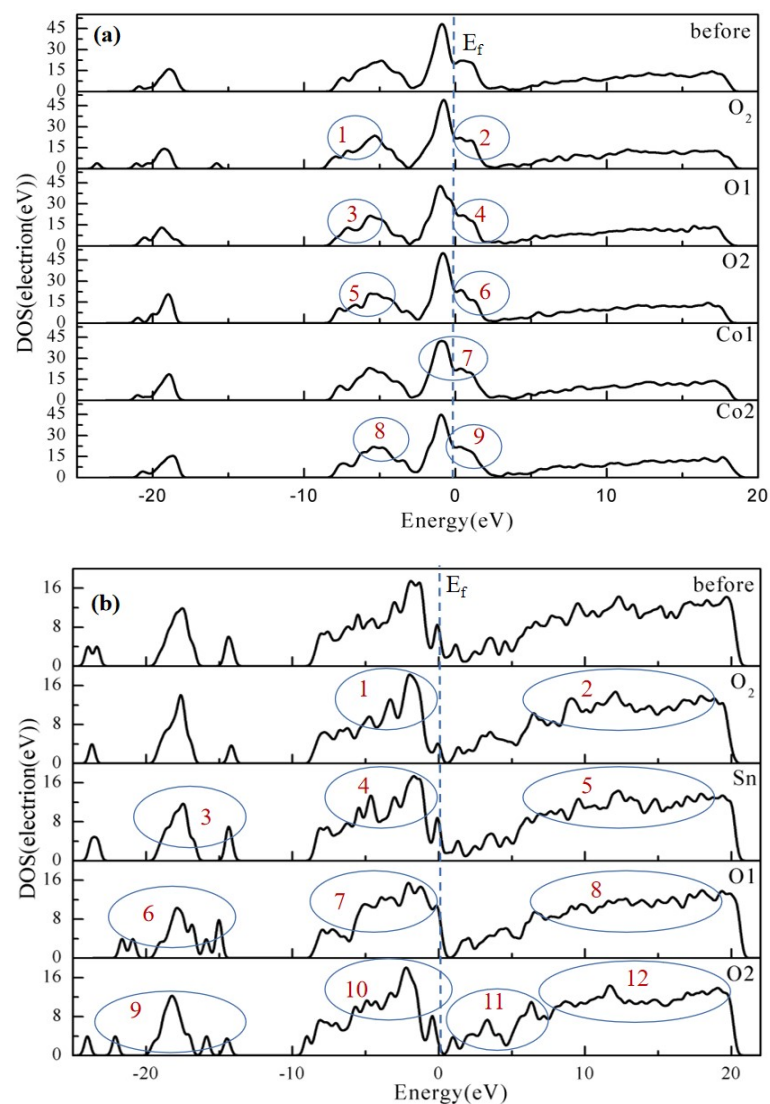
Figure S5. The DOS of H_2 adsorption on different sites of CoO(110)(II) and SnO$_2$(100)(II) surface. **(a)** H_2 on O$_2$ molecule, and O1 site, O2 site, Co1 site, Co2 site of CoO(110)(II) surface. **(b)** H_2 on O$_2$ molecule, and O1 site, O2 site, Sn site of SnO$_2$(100)(II) surface.

Figure S5(a) shows the DOS that H_2 is adsorbed on CoO(110)(II) surface. The DOS value is reduced when H_2 is adsorbed on O site of O$_2$ molecule as labeled 1 and 2. It implies the electronic resistance of CoO is enlarged. When H_2 is adsorbed on O1 site, DOS has obvious changes at Fermi level. The peaks near Fermi level are obviously disappear and the DOS value are decreased as labeled 3 and 4, indicated the CoO electronic resistance is increased. When H_2 is adsorbed on O2 site, the DOS

value has obvious reduce as labeled 5 and 6. Consequently, the CoO(110) electronic resistance is enhanced when H_2 is adsorbed on O2 site. The DOS value is significantly lower at Fermi level as labeled 7 when H_2 is adsorbed on Co1 site. It reflects the increases of electronic resistance for the adsorption of H_2. When H_2 is adsorbed on Co2 site, the changes of DOS occurs at the Fermi level as labeled 8 and 9. The DOS value at the Fermi level has the remarkable reduction, revealing the CoO electronic resistance is increased. On the whole, the CoO electrical resistance is increased when H_2 is adsorbed on different sites of the surface.

Figure S5(b) shows the DOS of H_2 adsorption on SnO_2(100)(II) surface. The DOS has evident changes when H_2 is adsorbed on O site of O_2 molecule. It indicates the SnO_2 electrical resistance has obvious changes. There are the formation of new peaks and increases of the DOS value (as labeled 1 and 2) when H_2 is adsorbed on Sn site. It implies the decrease of SnO_2 electronic resistance. DOS value has slight increases at Fermi level and amount of peaks are appear (as labeled 3, 4 and 5) when H_2 is adsorbed on Sn site. It reveals the enlargement of electrical conductivity. The DOS value has apparent raise (as labeled 6, 7 and 8) when H_2 is adsorbed on O1 site. It proves the decrease of SnO_2(100) electronic resistance. There are the formation of new peaks and the DOS value also has distinct enlargement, when H_2 is adsorbed O2 site. Therefore, SnO_2(100) electrical conductivity is enhanced when H_2 is adsorbed on O2 site. According to the above analysis, the SnO_2 resistance is reduced with H_2 adsorption on different sites of the surface.