Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Band alignment in CdS-α-Te van der Waals heterostructures for photocatalytic applications: Influence of biaxial strain and electric field

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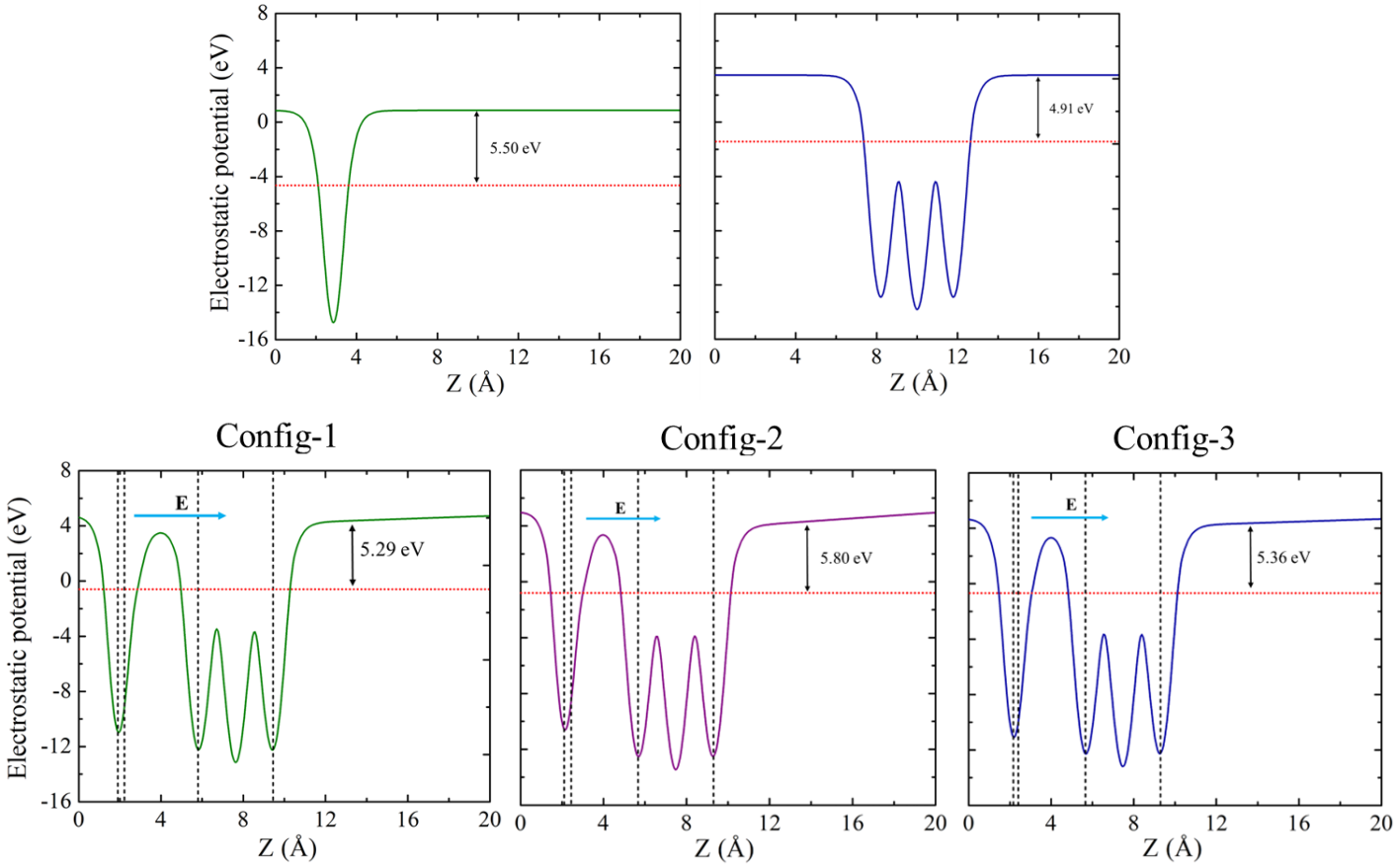
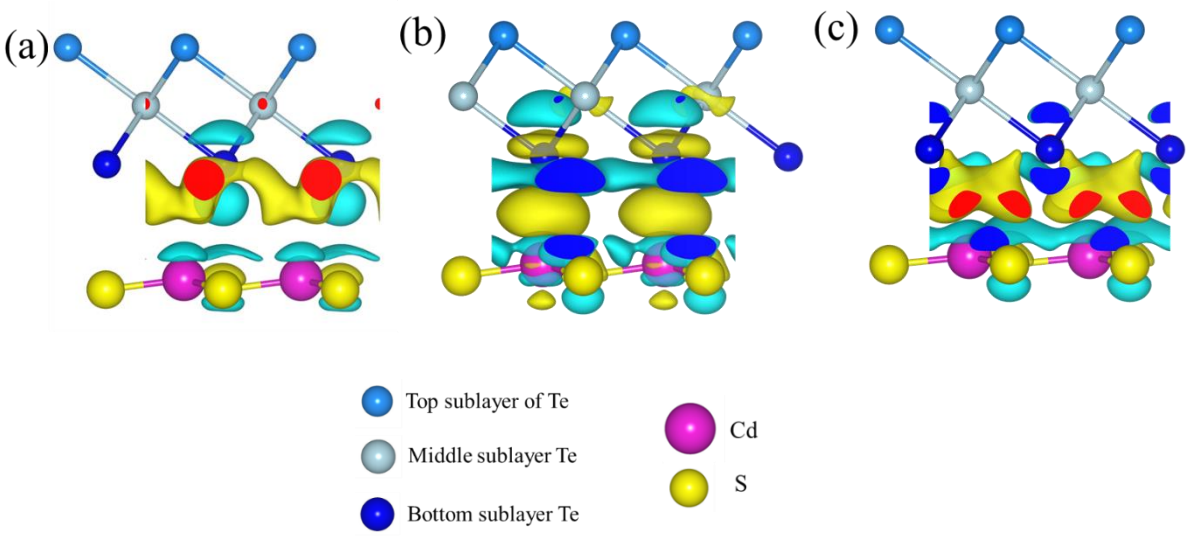
Supplementary Information



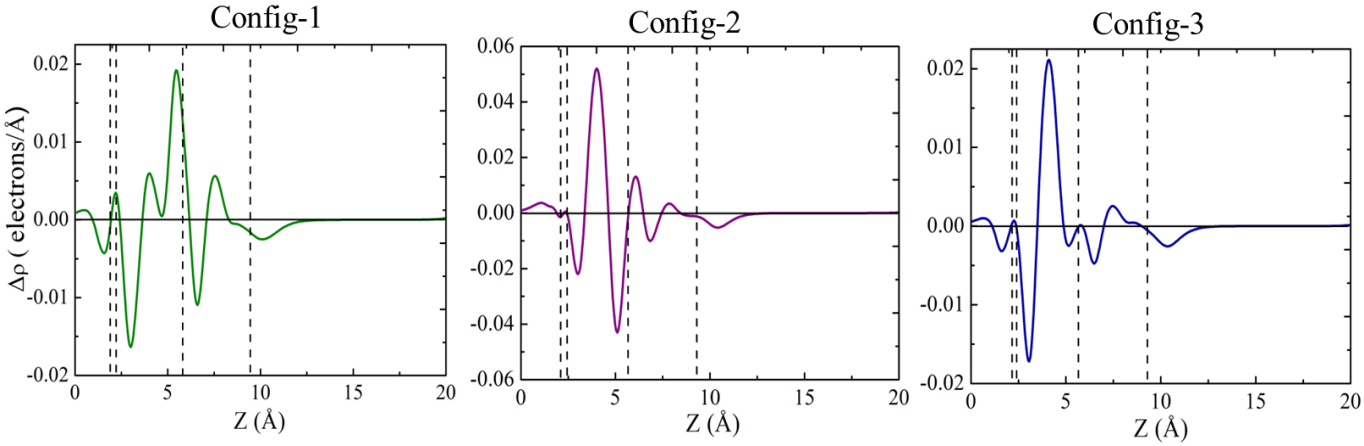
SFig. 1: The electrostatic potential of the CdS, α-Te SLs and the three configurations of the CdS- α-Te heterostructure. The vertical dashed lines represent the boundaries of the CdS (left) and α-Te (right) layers. The light blue arrow denotes the direction of the built-in electric field E. The red dotted line represents the Fermi-level. The results presented here are within the GGA approximation.

STable 1: Vacuum level and work functions of CdS and α-Te SLs and CdS-α-Te heterostructure.

Structure	**Vacuum level (eV)**	**Work function (eV)**
CdS	0.86	5.50
α-Te	3.44	4.91
Config-1	4.71	5.29
Config-2	4.98	5.80
Config-3	4.68	5.36



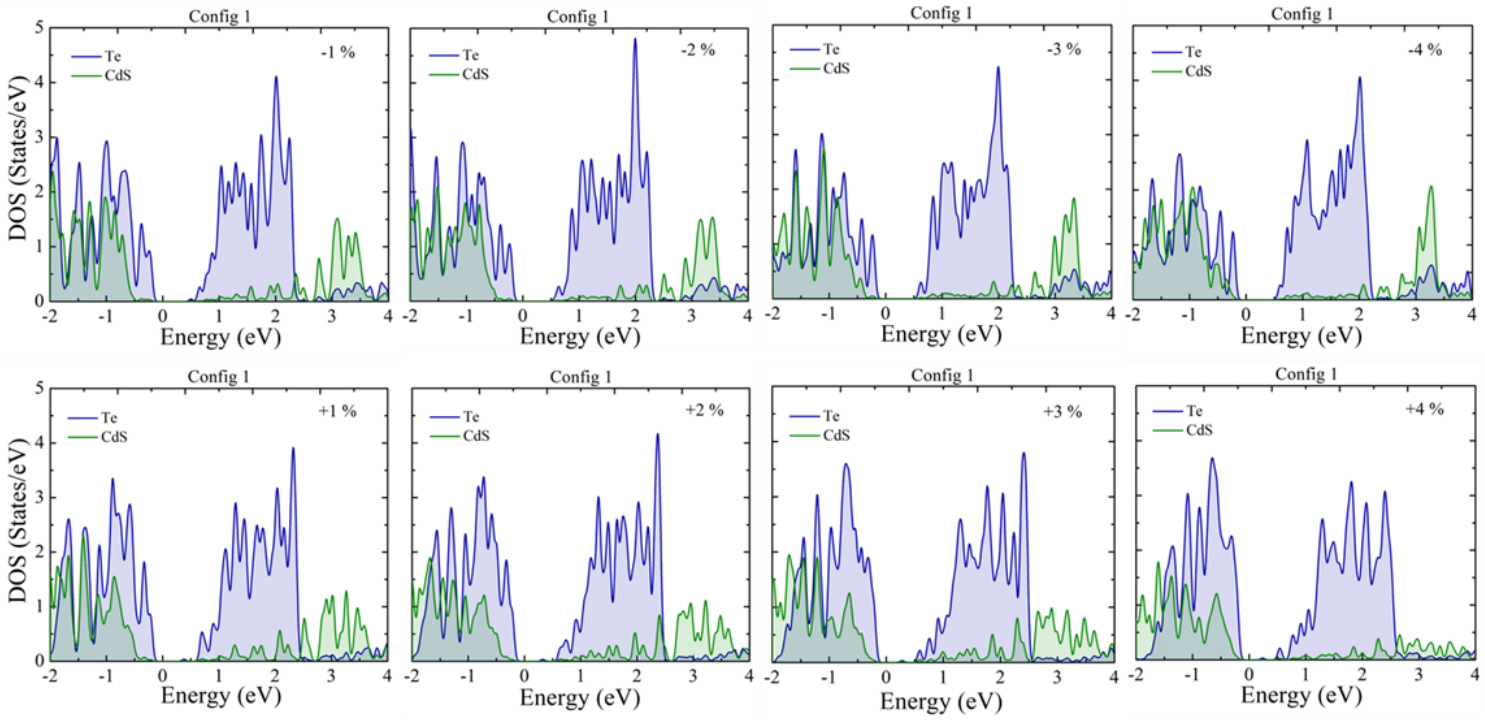
SFig. 2: Differential electron density representation in (a) config-1, (b) config-2 and (c) config-3 of the CdS-α-Te heterostructure with isosurface level 0.0002 e/Å^3. The yellow and cyan colored regions around the atoms represents the electron accumulation and depletion respectively. Note, that the red and dark blue areas solely denote cross-sections of yellow and cyan regions with the plane of the picture.

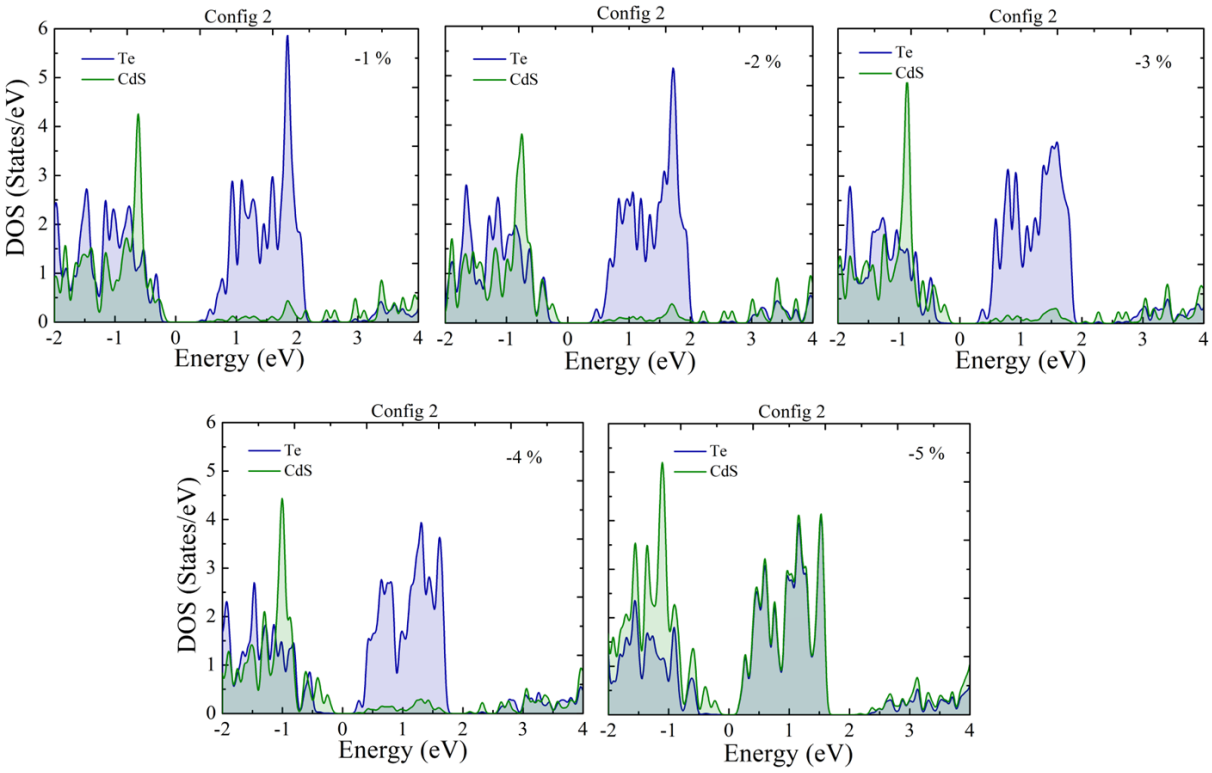


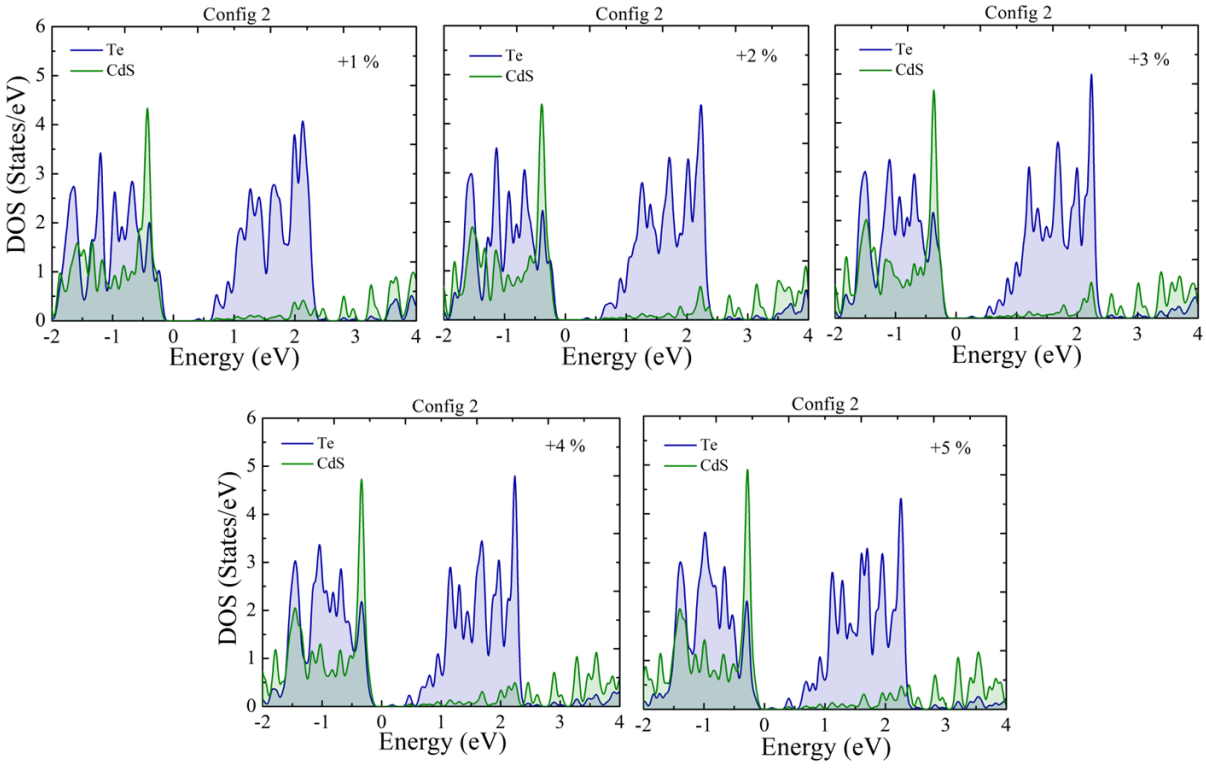
SFig. 3: Planar averaged electron density difference in the three configurations of the CdS-α Te heterostructure. Vertical dashed lines mark the CdS and α-Te regions. The positive and negative values represent the electron accumulation and depletion respectively.

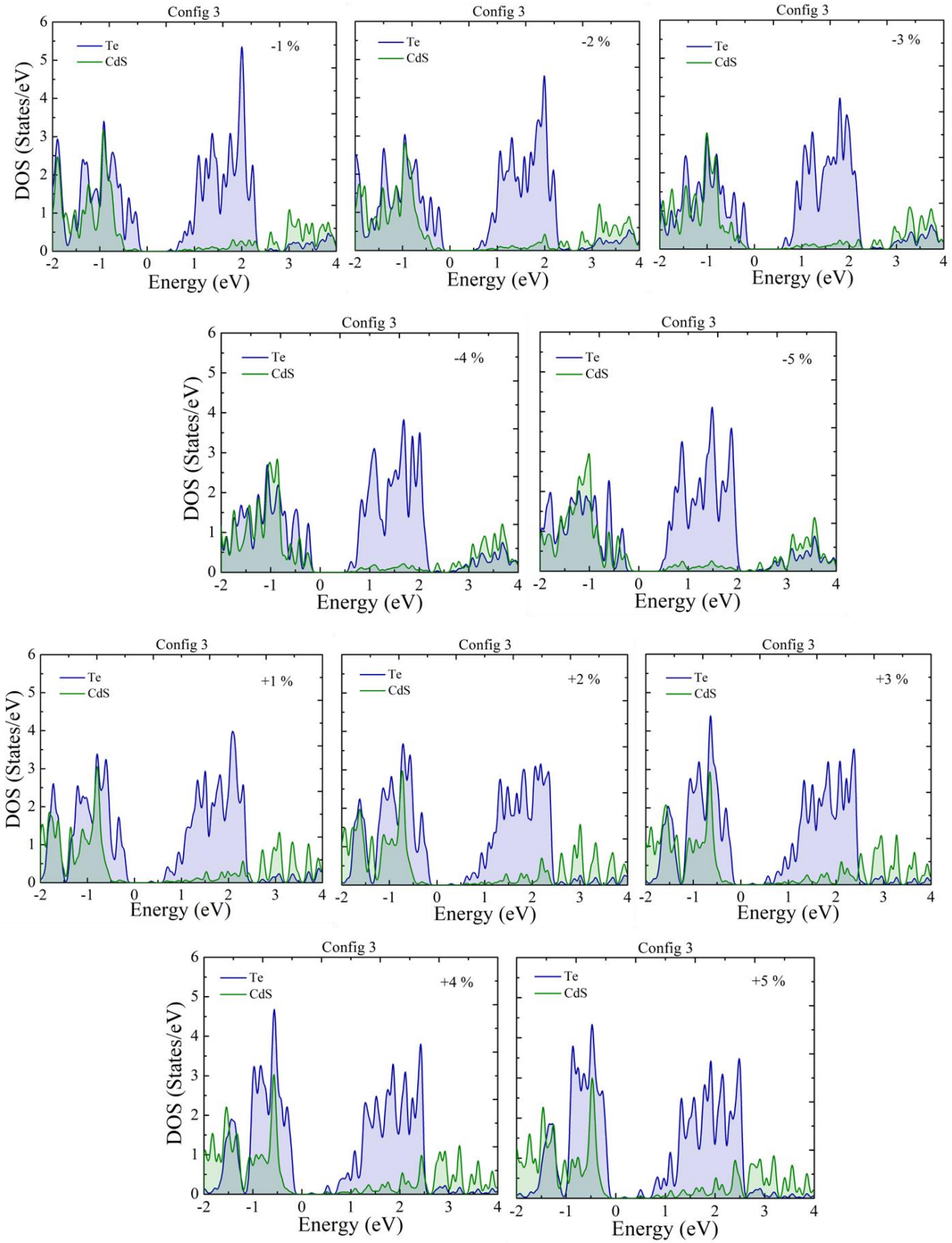
STable 3: Interlayer distance variation with reference to applied biaxial strain in the three configurations of the CdS- α-Te heterostructure.

Strain (%)	Interlayer distance (Å)		
	Config-1	Config-2	Config-3
-5	-	3.00	3.10
-4	3.43	3.08	3.13
-3	3.47	3.14	3.16
-2	3.51	3.19	3.18
-1	3.55	3.23	3.21
0	3.59	3.23	3.25
1	3.62	3.27	3.24
2	3.65	3.26	3.25
3	3.67	3.25	3.24
4	3.69	3.23	3.22
5	-	3.20	3.20

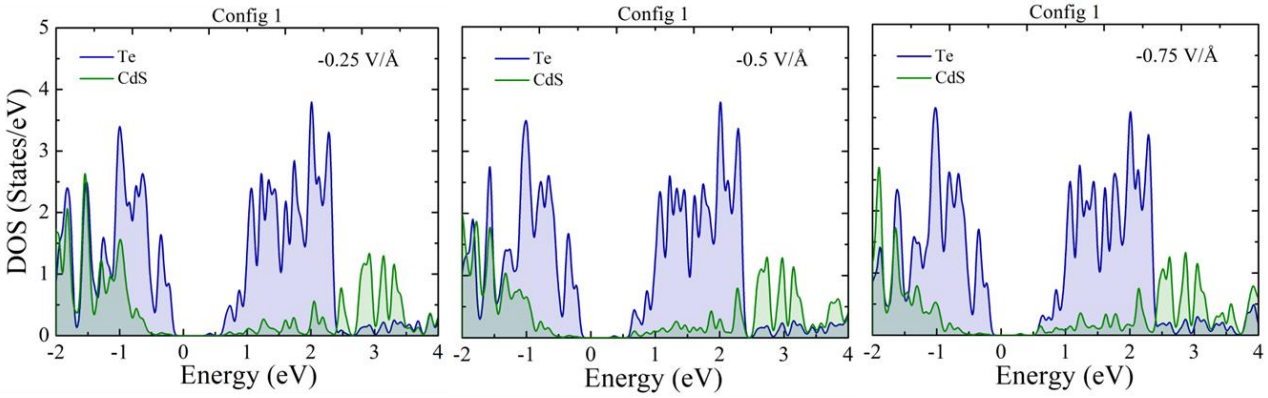



Config 2
Te
CdS
-1 %
-2 %
-3 %
-4 %
-5 %
DOS (States/eV)
Energy (eV)


Config 2
Te
CdS
+1 %
+2 %
+3 %
+4 %
+5 %
DOS (States/eV)
Energy (eV)



SFig 4: The evolution of layer resolved density (LDOS) of states of the three configurations of the CdS-α-Te heterostructure with respect to applied biaxial strain. The negative and positive strain values represent the compression and tension respectively. The Fermi-level is set to zero.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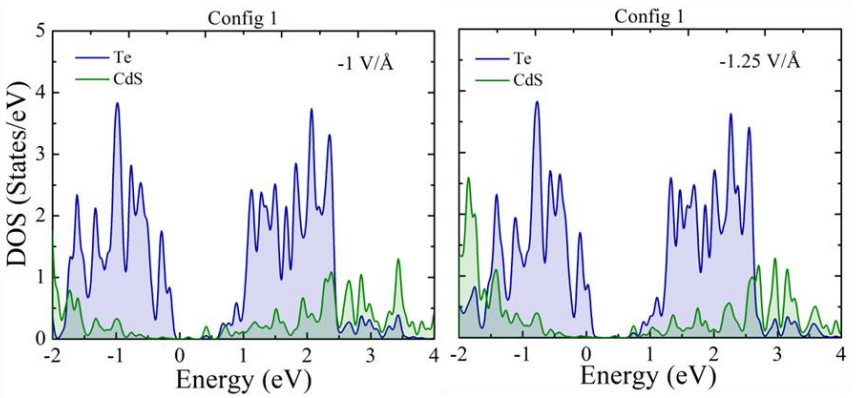
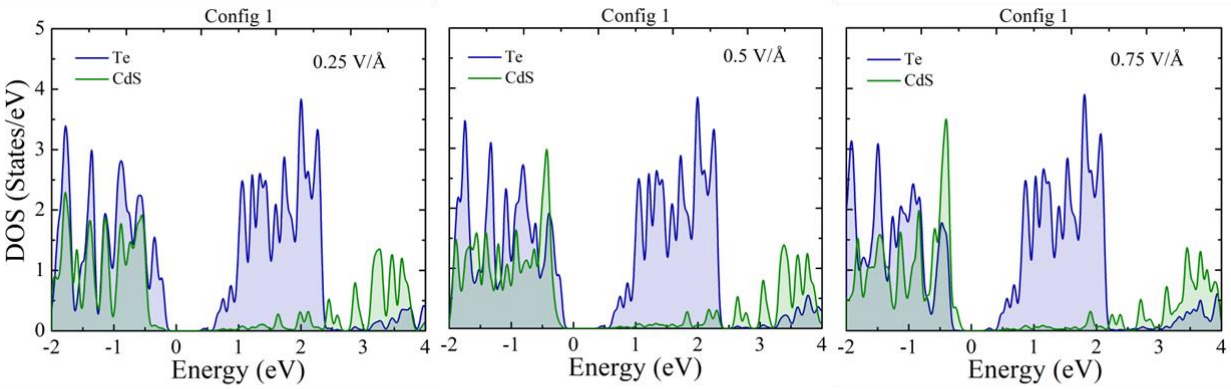
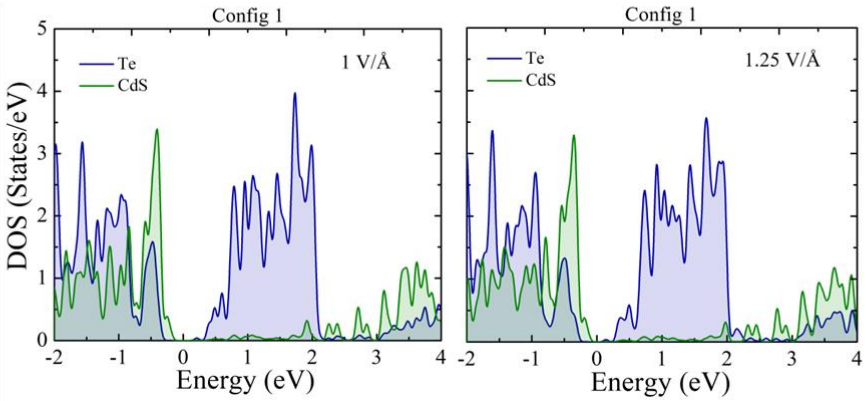
Te
CdS
-0.25 V/Å
-0.5 V/Å
-0.75 V/Å
DOS (States/eV)
Energy (eV)


Config 1
Te
CdS
-1 V/Å
-1.25 V/Å
DOS (States/eV)
Energy (eV)


Config 1
Te
CdS
0.25 V/Å
0.5 V/Å
0.75 V/Å
DOS (States/eV)
Energy (eV)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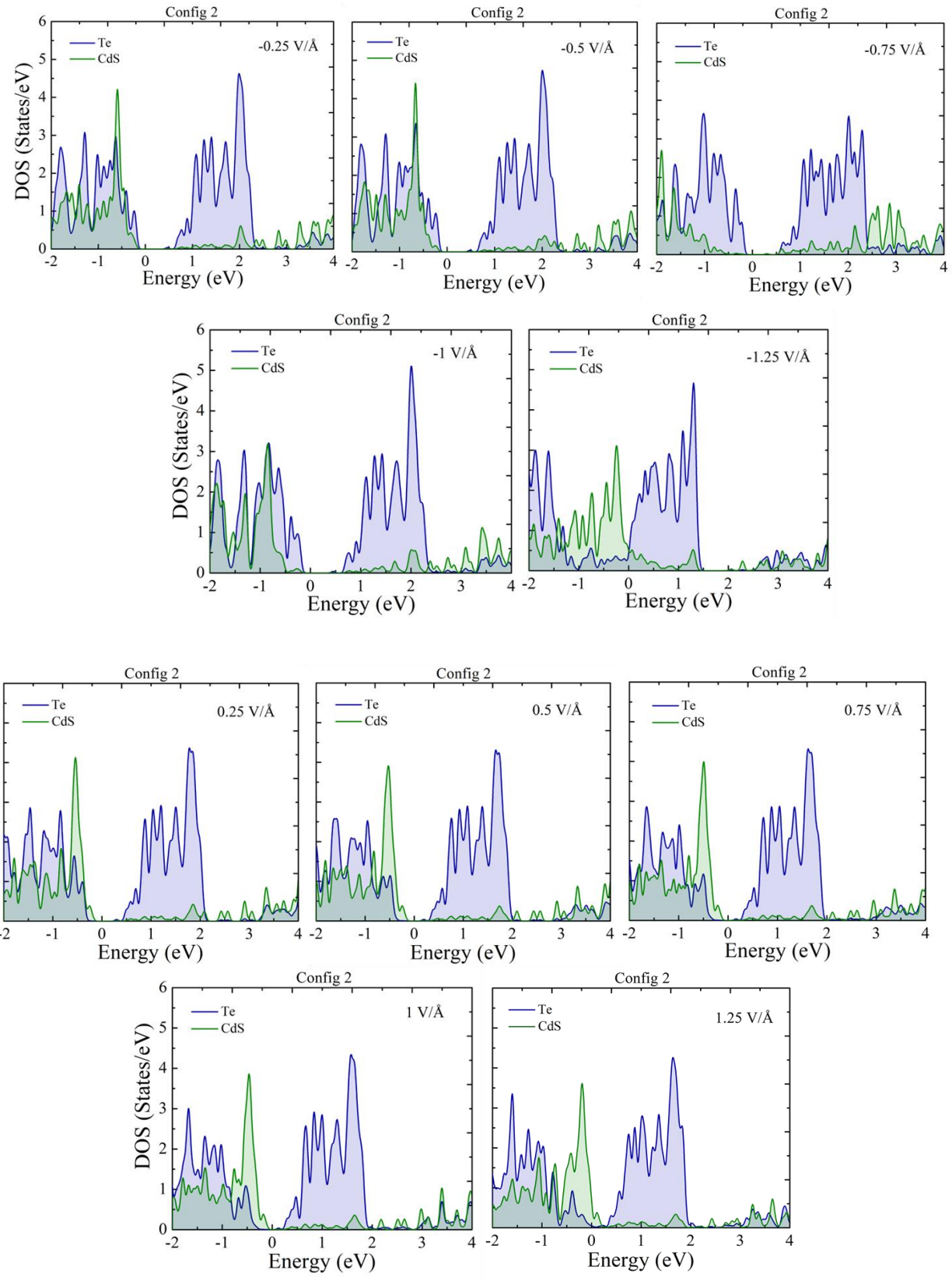
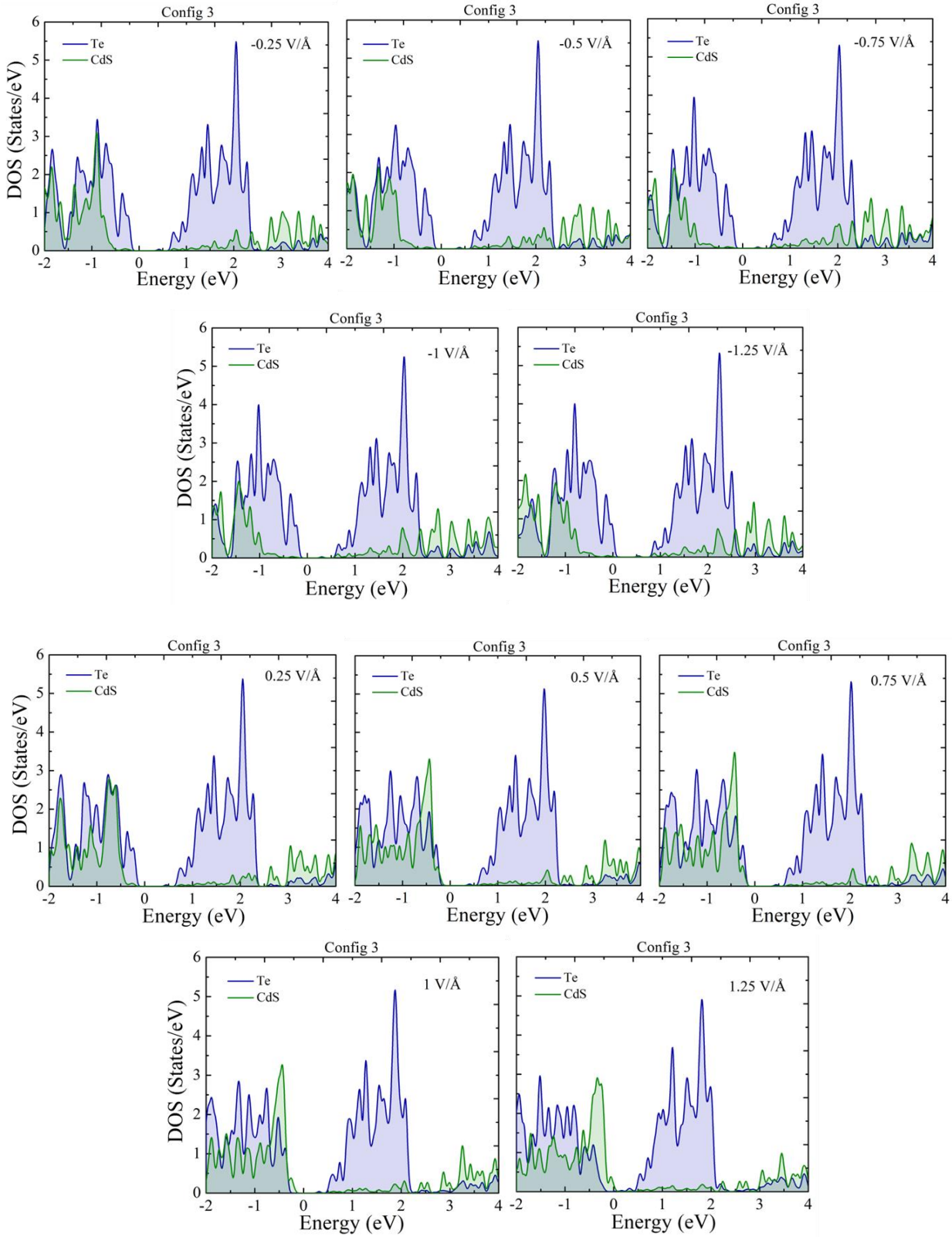
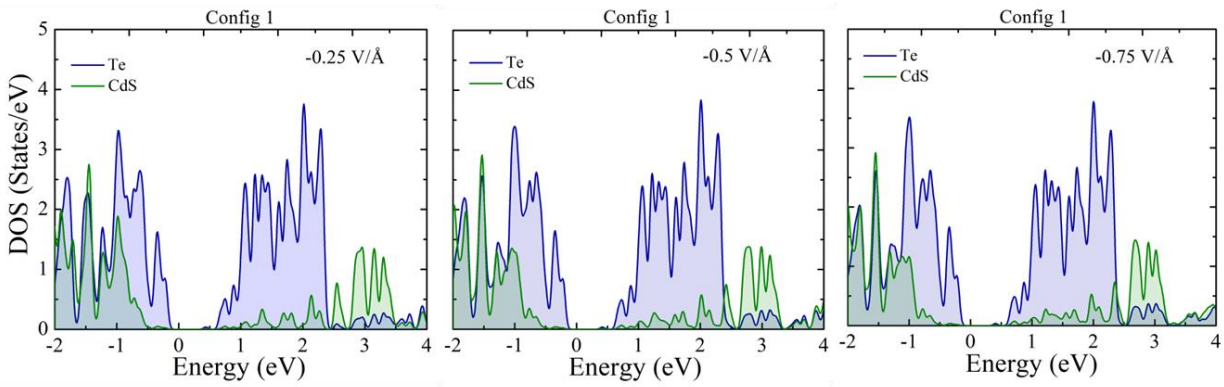
Te
CdS
1 V/Å
1.25 V/Å
DOS (States/eV)
Energy (eV)


Config 2
Te
CdS
-0.25 V/Å
-0.5 V/Å
-0.75 V/Å
-1 V/Å
-1.25 V/Å
0.25 V/Å
0.5 V/Å
0.75 V/Å
1 V/Å
1.25 V/Å
DOS (States/eV)
Energy (eV)



SFig. 5: The evolution of layer resolved density of states of the three configurations of the CdS-α-Te interface with respect to applied electric field. The negative and positive electric field values represent the direction of the fields pointing along negative and positive z-axis. respectively. The Fermi-level is set to zero. These are the results when the ionic relaxation is not performed.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Te
CdS
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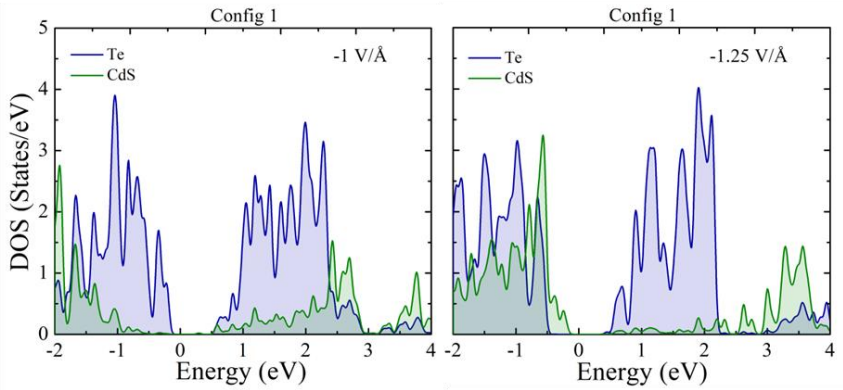
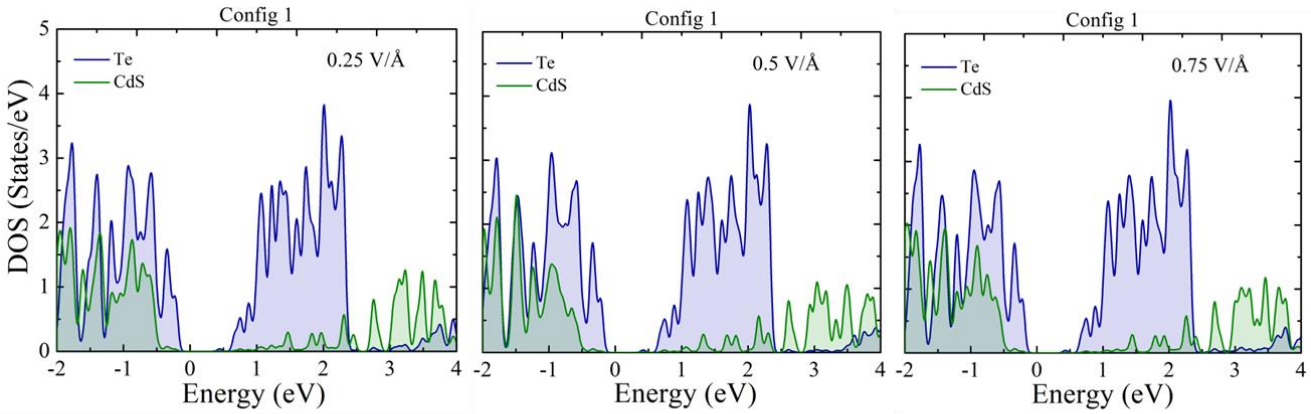
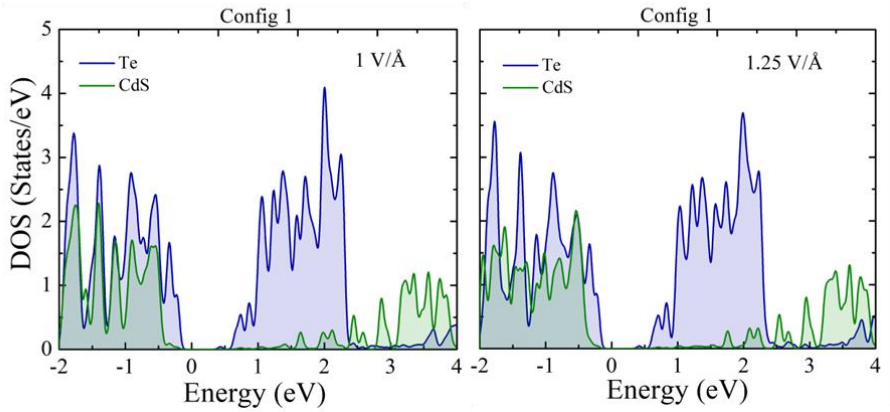
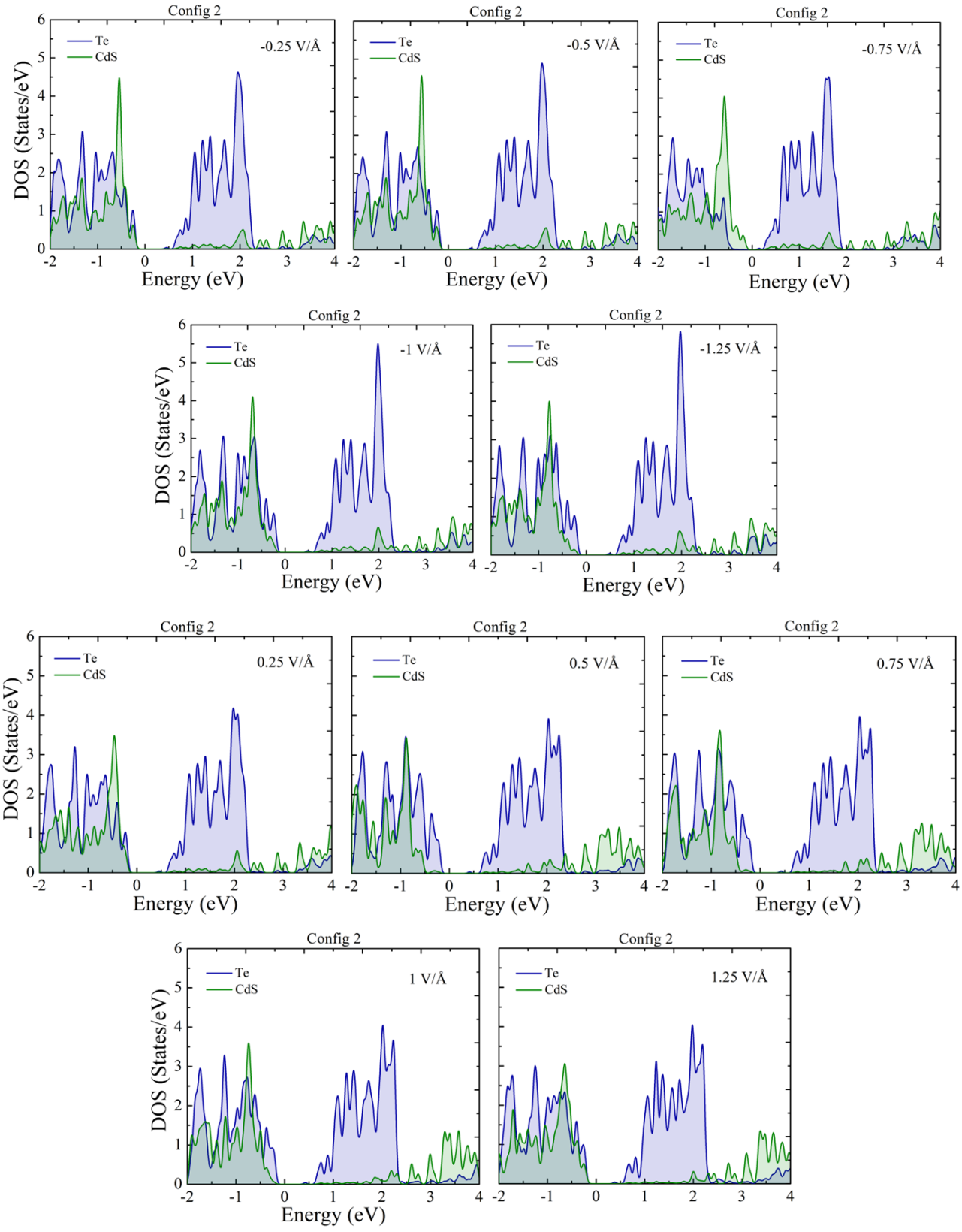
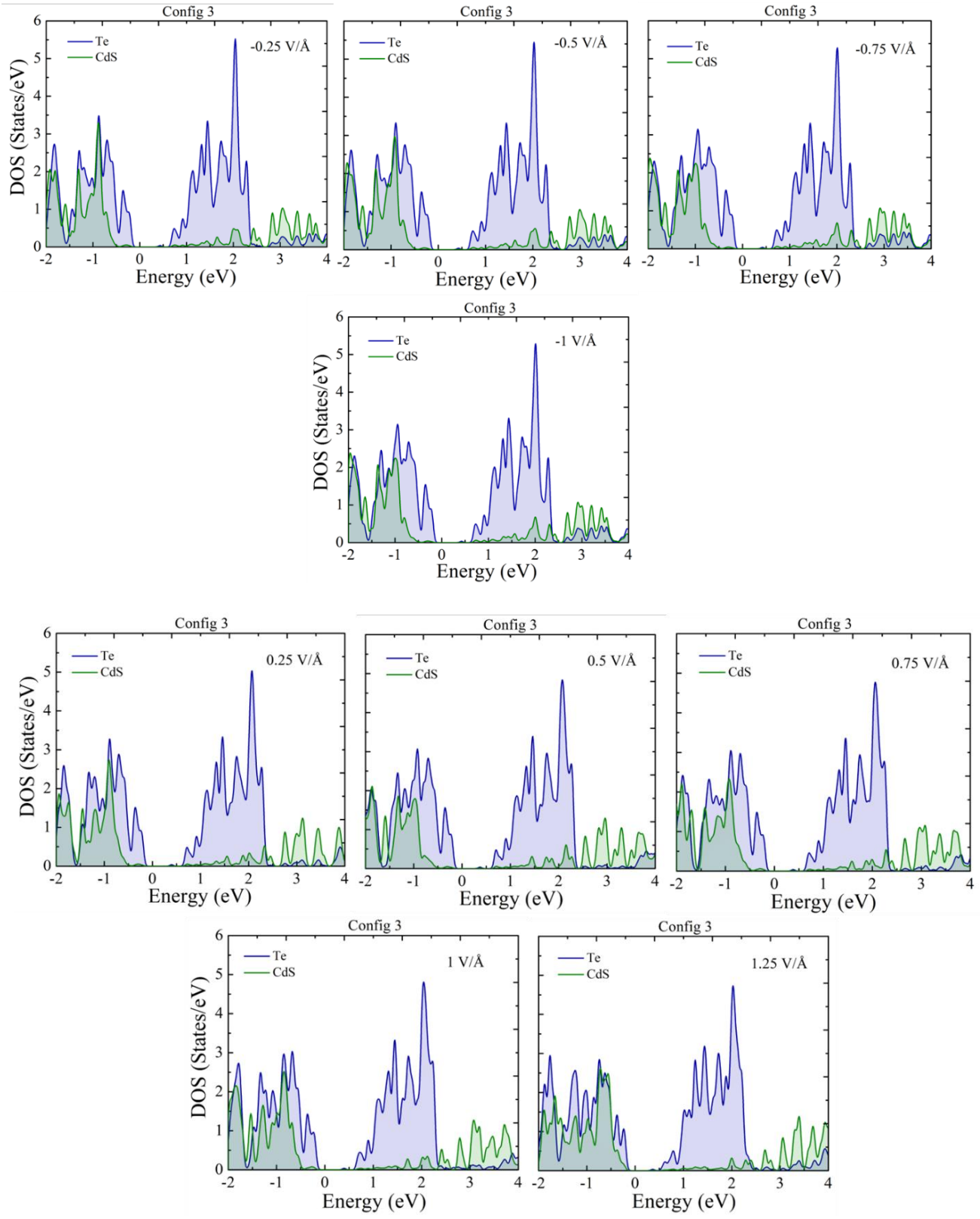
-0.5 V/Å
-0.75 V/Å
DOS (States/eV)
Energy (eV)


Config 1
Te
CdS
-1 V/Å
-1.25 V/Å
DOS (States/eV)
Energy (eV)


Config 1
Te
CdS
0.25 V/Å
0.5 V/Å
0.75 V/Å
DOS (States/eV)
Energy (eV)


Config 1
Te
CdS
1 V/Å
1.25 V/Å
DOS (States/eV)
Energy (eV)


Config 2
Te
CdS
-0.25 V/Å
-0.5 V/Å
-0.75 V/Å
-1 V/Å
-1.25 V/Å
0.25 V/Å
0.5 V/Å
0.75 V/Å
1 V/Å
1.25 V/Å
DOS (States/eV)
Energy (eV)



SFig. 6: The evolution of layer resolved density of states of the CdS-α-Te heterostructure with respect to applied electric field. The negative and positive electric field values represent the direction of the fields pointing along negative and positive z-axis. respectively. The Fermi-level is set to zero. These are the results when the ionic relaxation is performed.