Insights into the vacancy behaviour at the interface of As-Sb lateral heterostructures

Lei Ao* and Zhihua Xiong*

Key Laboratory for Optoelectronics and Communication of Jiangxi Province, Jiangxi Science & Technology

Normal University, Nanchang 330018, China

Email: al20060608@126.com

Email: xiong_zhihua@126.com

Table S1 Overview of the defect formation energy (E_f) and the electronic/magnetic properties including the total magnetic moment (Mag.), bandgap (E_g), orbital compositions of band edges of defective arsenene, antimonene, and As-Sb LHSs. The results for the pristine arsenene, antimonene, and As-Sb LHS are also given for comparison. The direct and indirect band gap are denoted as "d" and "i", respectively. Both of E_f and E_g are in the unit of eV. Mag. is in the unit of μ_B. E_f of the lowest-energy configuration in each group is italicized.

Defect	E_f	Mag.	E_g	VBM Orbital	CBM Orbital	Defect	E_f	Mag.	E_g	VBM Orbital	CBM Orbital
				Arsenene (As)						**Antimonene (Sb)**	
Pristine	-	-	1.61 (i)	As $(p_x + p_y)$	As $(p_y + p_z)$	Pristine	-	-	1.26 (i)	Sb $(p_x + p_y)$	Sb $(p_y + p_z)$
SV-59	*2.02*	1.00	0.56 (i) ↑	As p_x ↑	As $(p_x + p_y + p_z)$ ↑	SV-59	*1.70*	1.00	0.45 (i) ↑	Sb p_x ↑	Sb $(p_x + p_y + p_z)$ ↑
			0.91 (i) ↓	As $(p_y + p_z)$ ↓	As $(p_x + p_z)$ ↓				0.71 (i) ↓	Sb p_y ↓	Sb $(p_x + p_z)$ ↓
DV-585	2.31	-	1.06 (i)	As p_x	As $(p_x + p_y + p_z)$	DV-585	1.94	-	0.88 (i)	Sb p_x	Sb $(p_x + p_y + p_z)$
DV-555777	*2.26*	-	1.43 (d)	As $(p_x + p_y + p_z)$	As $(p_x + p_y + p_z)$	DV-555777	*1.89*	-	1.21 (d)	Sb $(p_x + p_y)$	Sb $(p_x + p_y + p_z)$
DV-55557777_1	2.66	-	1.40 (d)	As $(p_x + p_y + p_z)$	As $(p_x + p_y + p_z)$	DV-55557777_1	2.24	-	1.19 (d)	Sb p_y	Sb $(p_x + p_y + p_z)$
DV-55557777_2	2.63	-	1.39 (d)	As $(p_x + p_y + p_z)$	As $(p_x + p_y + p_z)$	DV-55557777_2	2.23	-	1.16 (d)	Sb p_y	Sb $(p_x + p_y + p_z)$
				As-Sb LHS							
Pristine	-	-	0.46	Sb p_y	As p_z	DV-3-585-o	1.78	1.76	0.55 (d) ↑	Sb p_y ↑	As p_z ↑
SV-As-1	1.30	0.33	Metal	-	-				Half-metal	-	-
SV-As-2	1.70	1.00	0.37 (i) ↑	Sb p_y ↑	As p_z↑	DV-3-555777_1	1.65	-	0.46 (i)	Sb p_y	As p_z
			0.14 (i) ↓	Sb p_y ↓	(As p_y + Sb p_y) ↓	DV-3-555777_2	1.60	-	0.48 (d)	Sb p_y	As p_z
SV-Sb-1	1.04	1.00	0.51 (d) ↑	Sb p_y ↑	As p_z↑	DV-3-55557777_1	1.67	-	0.53 (d)	Sb p_y	As p_z
			0.07 (d) ↓	Sb p_y ↓	As $(p_x + p_z)$ ↓	DV-3-55557777_2	1.58	-	0.55 (d)	Sb p_y	As p_z
SV-Sb-2	*0.98*	1.00	0.51 (d) ↑	Sb p_y ↑	As p_z↑	DV-3-55557777_3	*1.51*	-	0.56 (d)	Sb p_y	As p_z
			0.07 (d) ↓	Sb p_y ↓	Sb $(p_x + p_z)$ ↓						
DV-1-585	1.67	-	0.43 (d)	Sb p_y	As p_z						
DV-1-555777_1	2.41	-	0.36 (d)	Sb p_y	As p_z	DV-4-585-o	2.68	1.61	0.21 (i) ↑	Sb p_y ↑	(As + Sb) $(p_y + p_z)$ ↑
DV-1-555777_2	*1.39*	-	0.50 (d)	Sb p_y	As p_z				Half-metal	-	-
DV-1-555777_3	2.38	-	0.20 (d)	Sb p_y	As $(p_y + p_z)$	DV-4-555777_1	2.46	-	0.37 (d)	Sb p_y	As p_z
DV-1-555777_4	1.46	-	0.50 (d)	Sb p_y	As $(p_x + p_z)$	DV-4-555777_2	2.45	-	0.32 (d)	Sb p_y	As p_z
DV-1-55557777_1	2.20	-	0.40 (d)	Sb p_y	As p_z	DV-4-55557777_1	1.78	-	0.50 (d)	Sb p_y	As $(p_x + p_z)$

|											
DV-1-55557777_2	2.36	-	0.40 (d)	Sb p_y	As p_z	DV-4-55557777_2	1.79	-	0.50 (d)	Sb p_y	As p_z
DV-1-55557777_3	2.27	-	0.28 (d)	Sb p_y	As p_z	DV-4-55557777_3	*1.75*	-	0.51 (d)	Sb p_y	As p_z
DV-1-55557777_4	2.36	-	0.30 (d)	Sb p_y	As p_z						
DV-2-585	1.66	-	0.41 (d)	Sb p_y	(Sb p_z + As p_z)						
DV-2-555777_1	1.89	-	0.46 (d)	Sb p_y	As p_z	DV-5-585	*0.39*	-	0.55 (d)	Sb p_y	As p_z
DV-2-555777_2	*0.98*	-	0.57 (d)	Sb p_y	As p_z	DV-5-555777_1	1.01	-	0.55 (d)	Sb p_y	As p_z
DV-2-555777_3	1.97	-	0.38 (d)	Sb p_y	As p_z	DV-5-555777_2	0.97	-	0.55 (d)	Sb p_y	(Sb p_z + As p_z)
DV-2-555777_4	1.10	-	0.55 (d)	Sb p_y	As p_z	DV-5-55557777_1	1.76	-	0.52 (d)	Sb p_y	As p_z
DV-2-55557777_1	1.32	-	0.51 (d)	Sb p_y	As p_z	DV-5-55557777_2	1.75	-	0.56 (d)	Sb p_y	As p_z
DV-2-55557777_2	1.54	-	0.49 (d)	Sb p_y	As p_z	DV-5-55557777_3	1.74	-	0.58 (d)	Sb p_y	As p_z
DV-2-55557777_3	1.45	-	0.44 (d)	Sb p_y	As p_z						
DV-2-55557777_4	1.61	-	0.43 (d)	Sb p_y	As p_z						
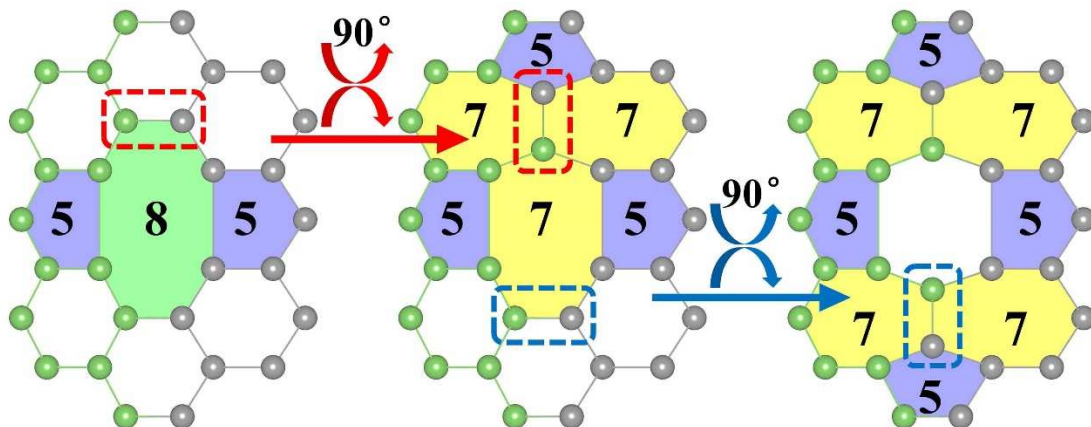
Fig. S1 Schematic view of the formation mechanisms of interface DV-555777s and DV-55557777s reconstructed from DV-585s. The pentagon, heptagon, and octagon are colored in blue, yellow, and green, respectively. The As and Sb atoms are represented by the green and gray balls, respectively.
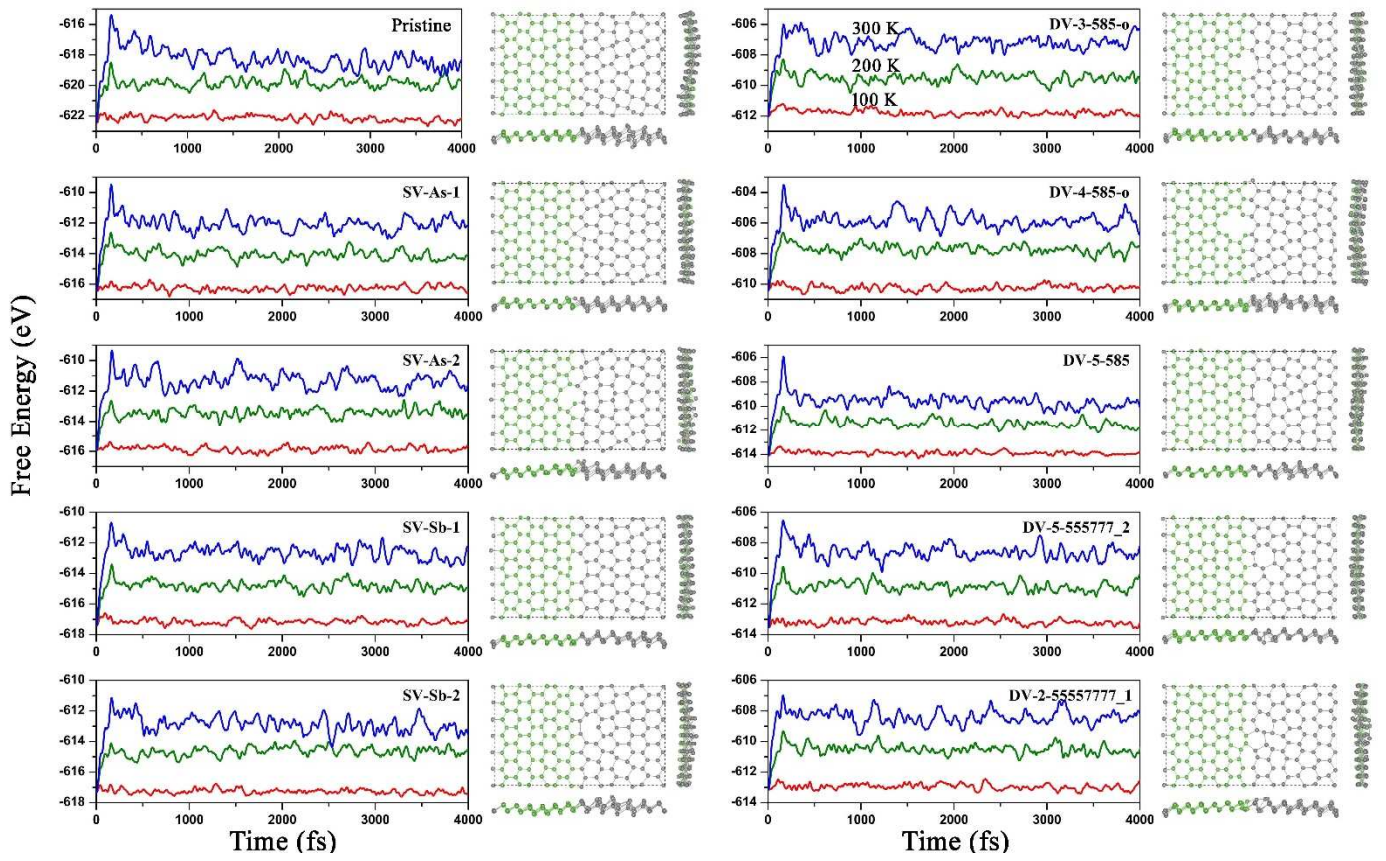
Fig. S2 Top (up) and side (down) views of the optimized structures of interface DVs in each group. The pentagon, heptagon, and octagon are colored in blue, yellow, and green, respectively. The As and Sb atoms are represented by the green and gray balls, respectively.

Fig. S3 The variation of the free energy (left) and the top/side views of the snapshot (right) for the pristine and defective As-Sb LHSs in the *ab initio* molecular dynamics simulation at 300 K during 4 ps.
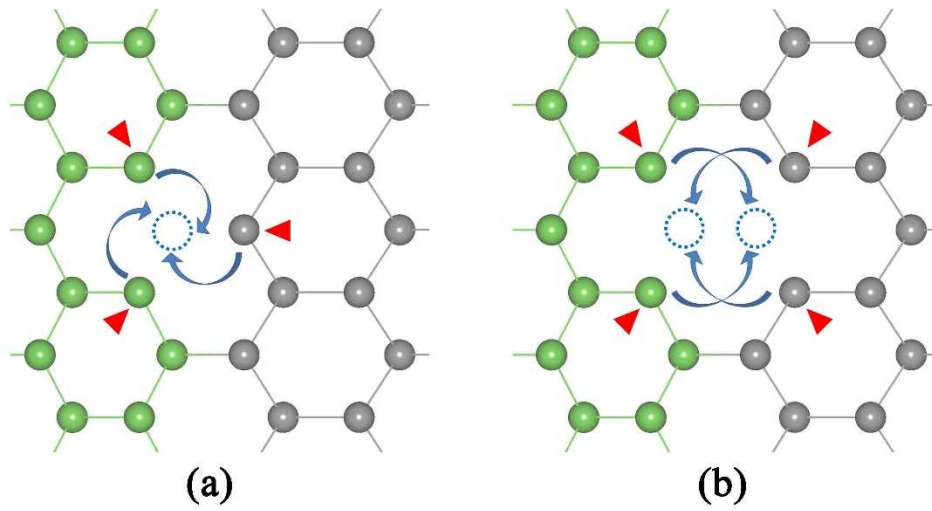
Fig. S4 The schematic view of the possible diffusion paths for the migration of interface (a) SVs and (b) DV-585s based on the atom-exchange mechanism. The atoms first-neighbouring to the vacant sites (denoted by dashed circles) are marked by red solid triangles. The As and Sb atoms are represented by the green and gray balls, respectively.
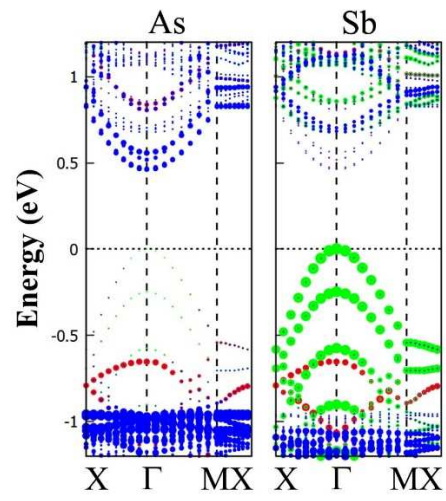
Fig. S5 The orbital-resolved band structure of pristine As-Sb LHS. The p_x, p_y, and p_z orbitals are shown by the red, green, and blue circles, respectively, where the size of the circle is proportional to the weight of the partial p orbital. The Fermi level is set at zero.
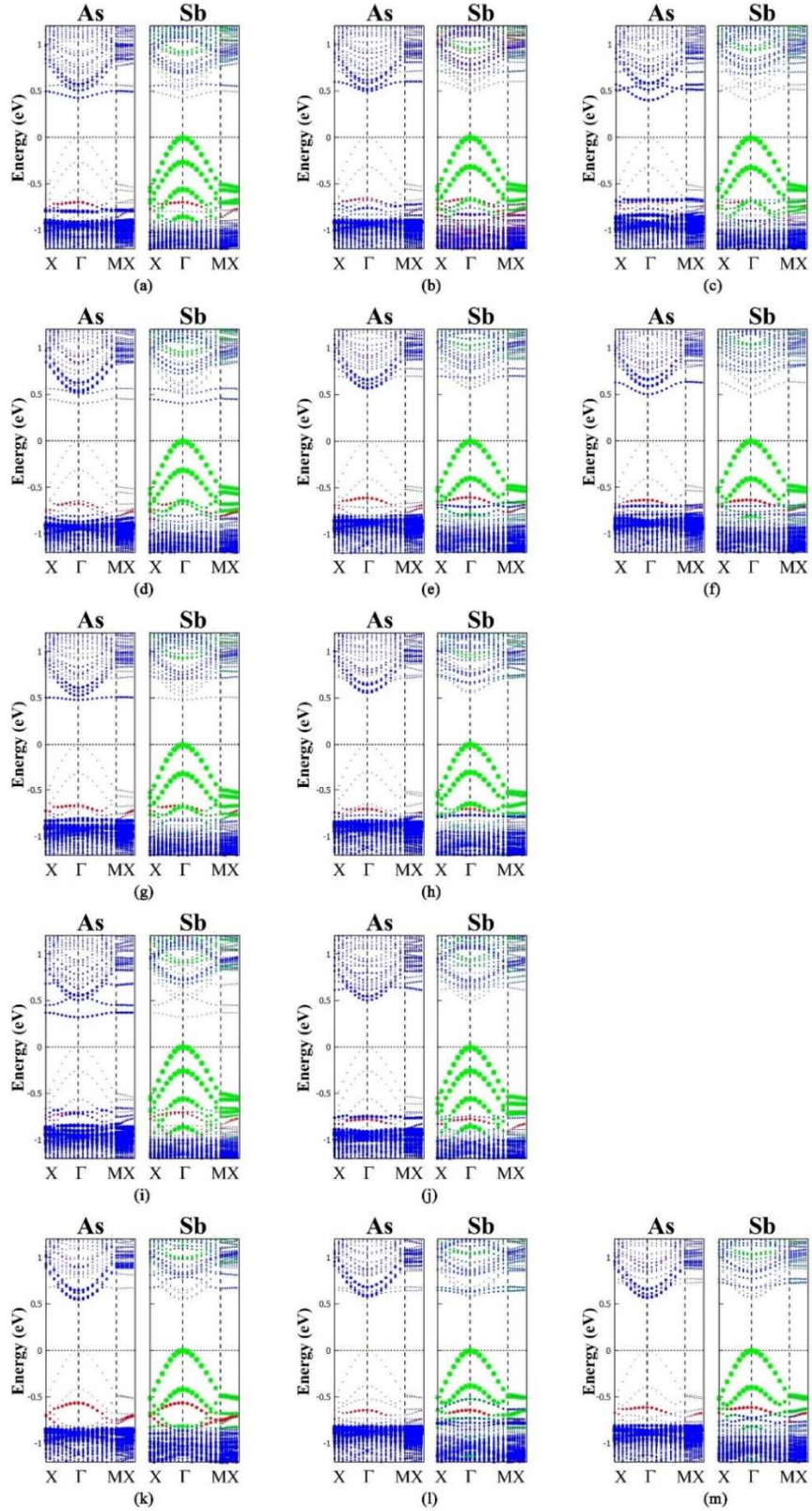
Fig. S6 The orbital-resolved band structures of As-Sb LHSs with nonmagnetic defects of (a) DV-1-585, (b) DV-1-555777_2, (c) DV-1-55557777_1, (d) DV-2-585, (e) DV-2-555777_2, (f) DV-2-55557777_1, (g) DV-3-555777_2, (h) DV-3-55557777_3, (i) DV-4-555777_2, (j) DV-4-55557777_3, (k) DV-5-585, (l) DV-5-555777_2 and (m) DV-5-55557777_3. The p_x, p_y, and p_z orbitals are shown by the red, green, and blue circles, respectively, where the size of the circle is proportional to the weight of the partial p orbital. The Fermi level is set at zero.
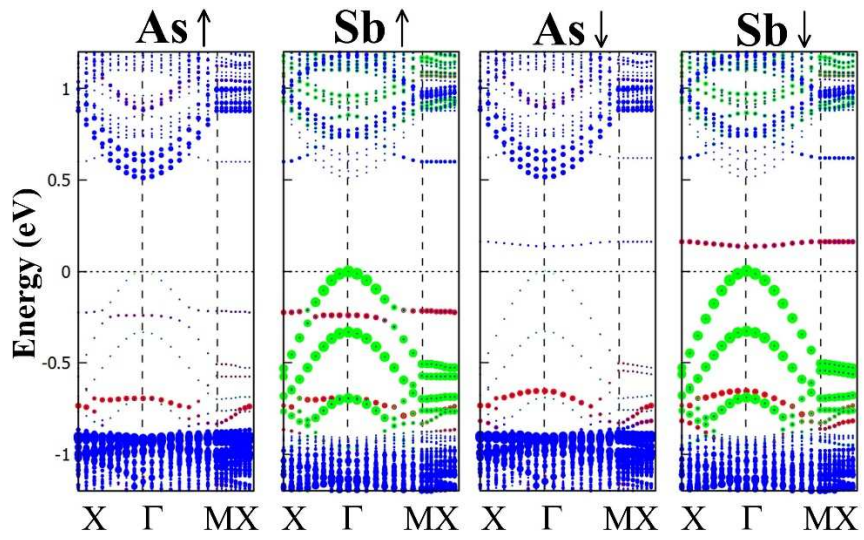
Fig. S7 The orbital-resolved band structure of defective As-Sb LHS with SV-1. Note that SV-1 is the final states in the migration process of SV-As-1 perpendicular to the interface as illustrated in Fig. 4(a). The p_x, p_y, and p_z orbitals are shown by the red, green, and blue circles, respectively, where the size of the circle is proportional to the weight of the partial p orbital. The Fermi level is set at zero.
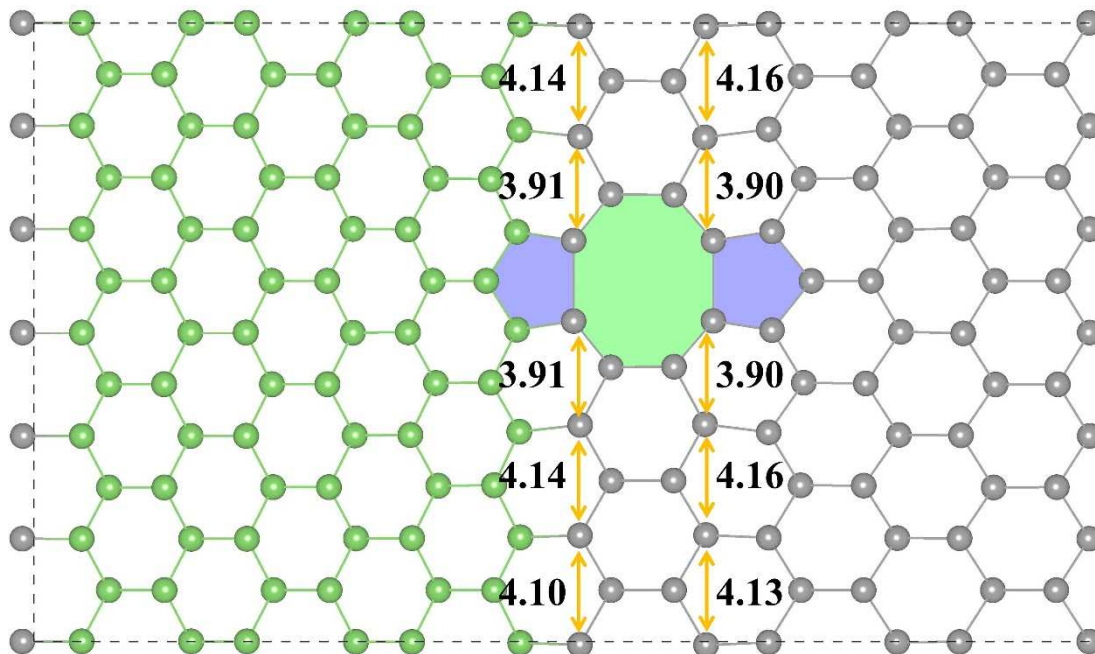
Fig. S8 Optimized Sb-Sb distances (in the unit of Å) parallel to the interface of DV-5-585 decorated As-Sb LHSs. The pentagon and octagon are colored in blue and green, respectively. The As and Sb atoms are represented by the green and gray balls, respectively.