Supporting Information for:

Blocking recombination centers by controlling the charge density of sulfur vacancy in antimony trisulfide

Xiao Han, Qi Zhao, Xiaodan, Yan, Ting Meng*, Jinlu He*

College of Chemistry and Chemical Engineering, Inner Mongolia University, Hohhot 010021, PR China

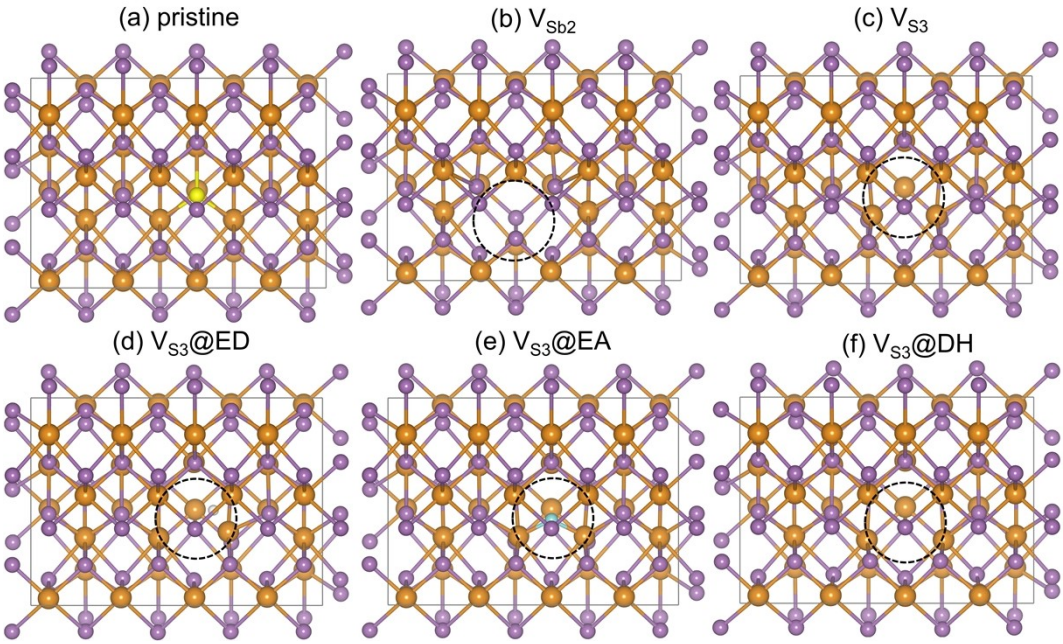

Fig. S1 Side views of optimized geometries for (a) pristine Sb_2S_3, (b) V_{Sb2}, (c) V_{S3}, (d) V_{S3}@ED, (e) V_{S3}@EA and (f) V_{S3}@DH systems. The Sb, S, O and H atoms are colored in orange, purple, light blue and white, respectively. The removed sulfur atom is colored in yellow for pristine Sb_2S_3 system. The dotted line circle denote the defect and elements doping region.

* Corresponding author, E-mail: t.meng@imu.edu.cn; hejinlu@imu.edu.cn.

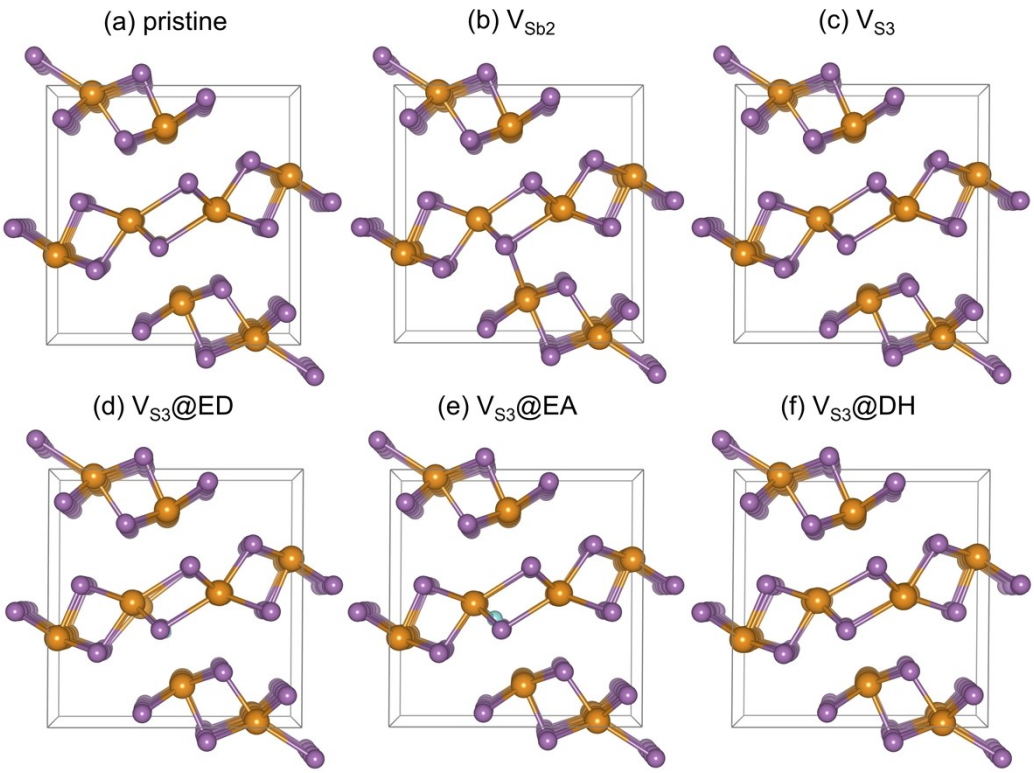

Fig. S2 Top views of optimized geometries for (a) pristine Sb_2S_3, (b) V_{Sb2}, (c) V_{S3}, (d) V_{S3}@ED, (e) V_{S3}@EA and (f) V_{S3}@DH systems. The Sb, S, O, and H atoms are colored in orange, purple, light blue and white, respectively. The removed sulfur atom is colored in yellow for pristine Sb_2S_3 system.

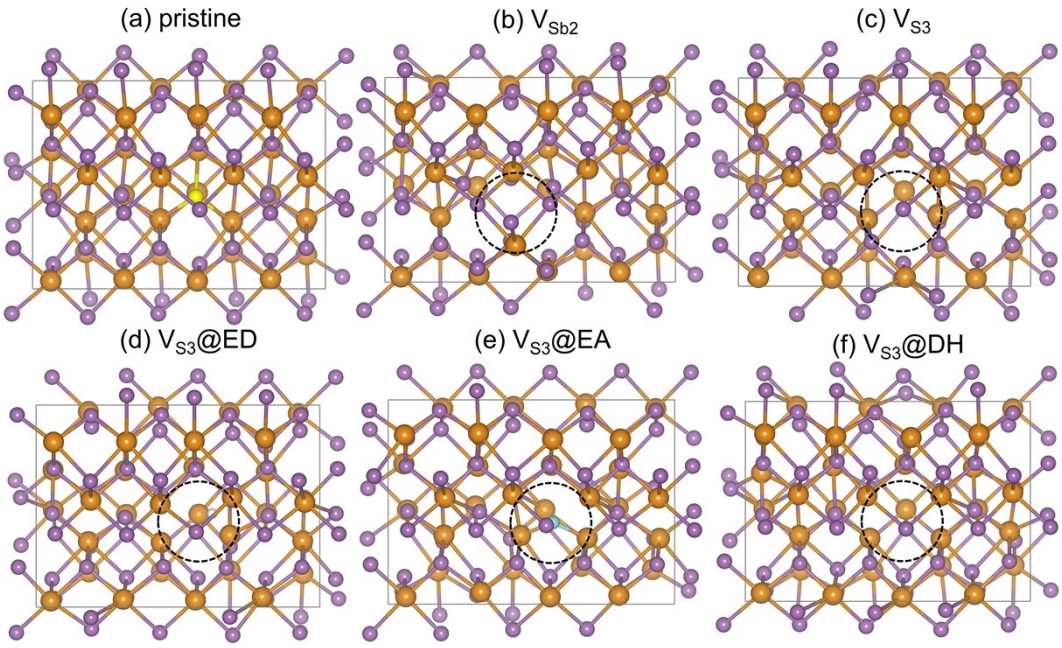

Fig. S3 Side views of representative geometries of 4 ps MD trajectories for (a) pristine Sb_2S_3, (b) V_{Sb2}, (c) V_{S3}, (d) V_{S3}@ED, (e) V_{S3}@EA and (f) V_{S3}@DH systems at 300 K. The Sb, S, O, and H atoms are colored in orange, purple, light blue and white, respectively. The removed sulfur atom is colored in yellow for pristine Sb_2S_3 system.

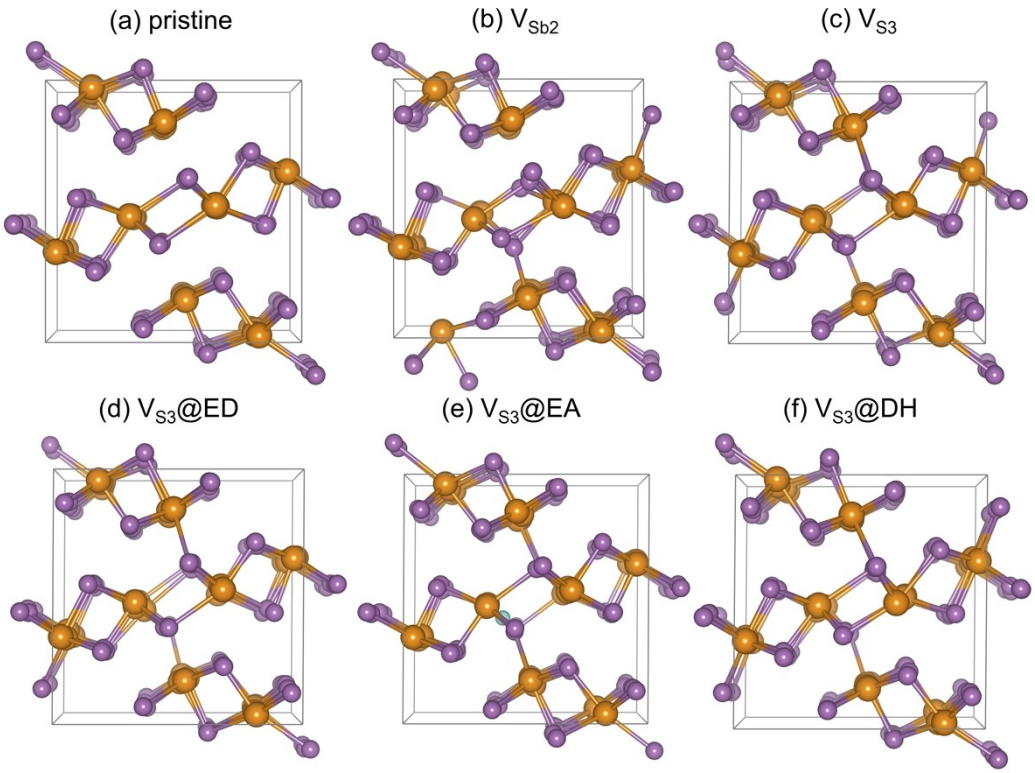

Fig. S4 Top views of representative geometries of 4 ps MD trajectories for pristine Sb_2S_3, V_{Sb2}, V_{S3}, V_{S3}@ED, V_{S3}@EA and V_{S3}@DH systems at 300 K. The Sb, S, O, and H atoms are colored in orange, purple, light blue and white, respectively. The removed sulfur atom is colored in yellow for pristine Sb_2S_3 system.

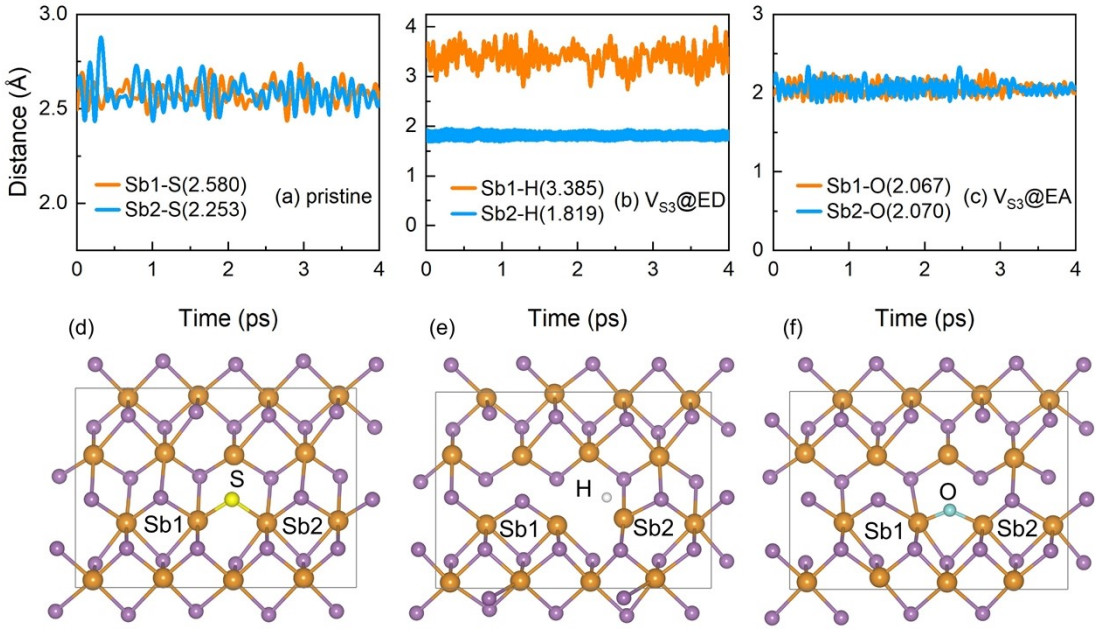

Fig. S5. Top panel shows evolution of the Sb-M (M = S, H and O) distances for pristine (a)Sb_2S_3, (b)V_{S3}@ED and (c)V_{S3}@EA systems. The bottom panel shows geometries for pristine (a)Sb_2S_3, (b)V_{S3}@ED and (c)V_{S3}@EA system. The numbers inside the parentheses are canonically averaged distance of Sb-M bonds. The Sb, S, O, and H atoms are colored in orange, purple, light blue and white, respectively. The removed sulfur atom is colored in yellow for pristine Sb_2S_3 system.

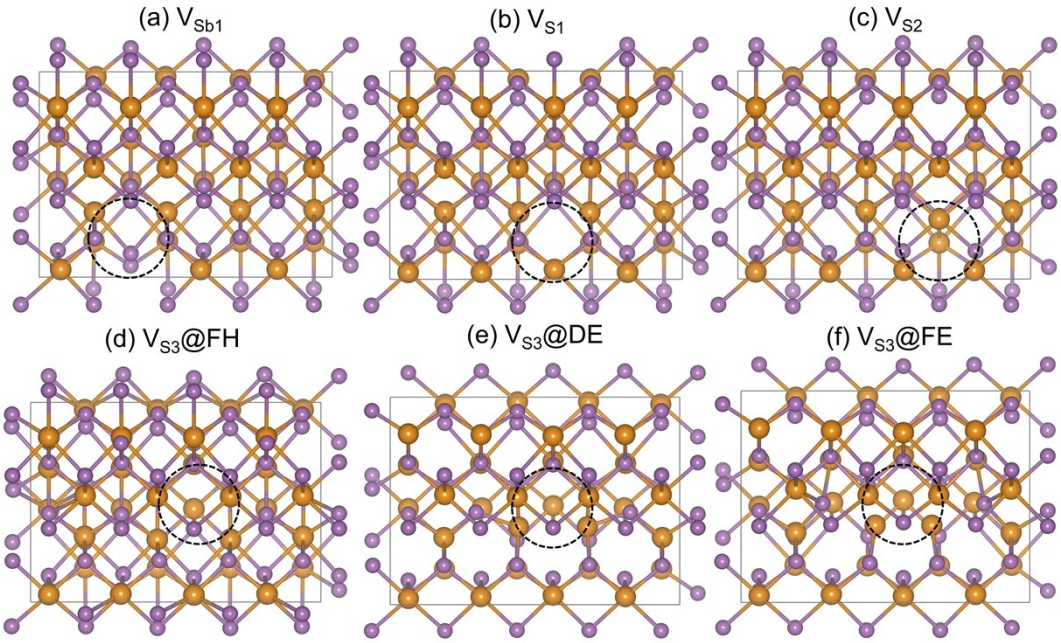

Fig. S6 Side views of optimized geometries for V_{Sb1}, V_{S1}, V_{S2}, V_{S3}@FH, V_{S3}@DE and V_{S3}@FE systems. The Sb and S atoms are colored in orange and purple, respectively.

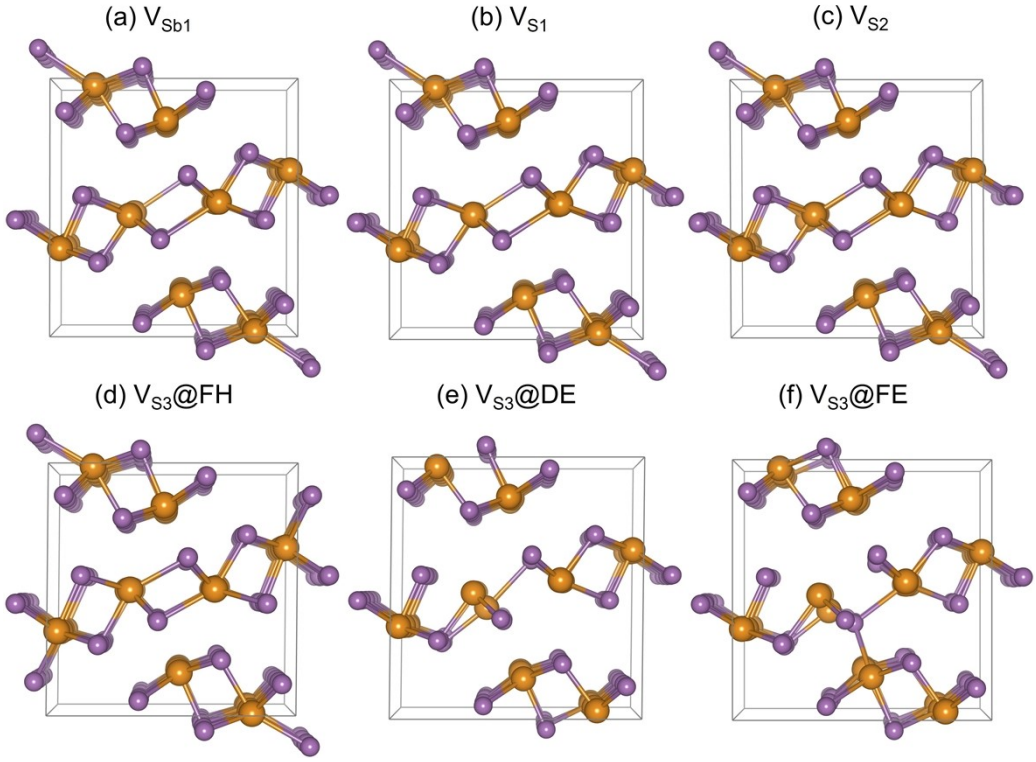

Fig. S7 Top views of optimized geometries for V_{Sb1}, V_{S1}, V_{S2}, V_{S3}@FH, V_{S3}@DE and V_{S3}@FE systems. The Sb and S atoms are colored in orange and purple, respectively.

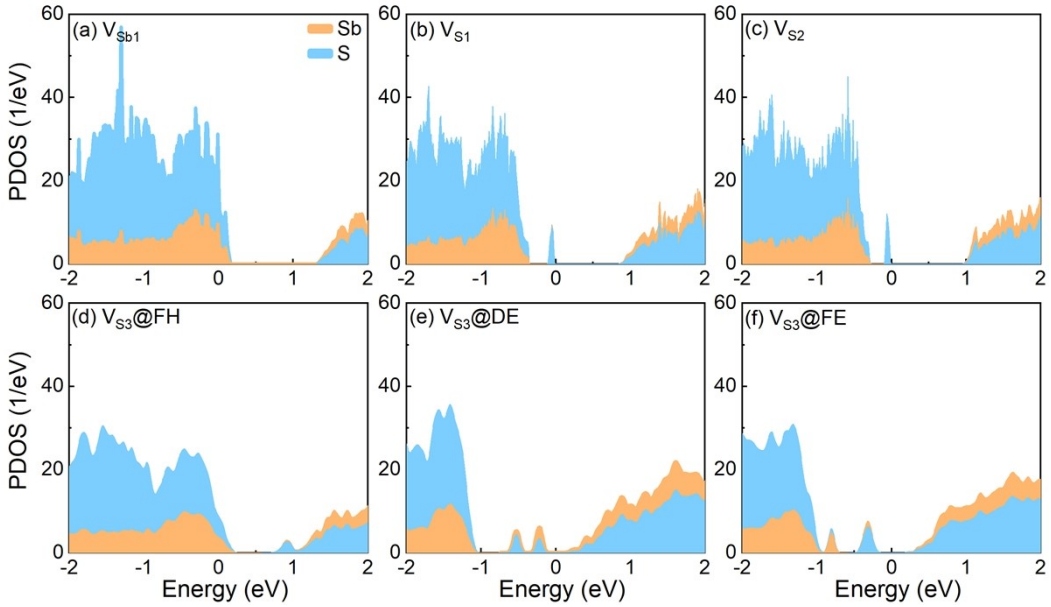

Fig. S8. Projected density of states (PDOS) of (a) V_{Sb1}, (b) V_{S1}, (c) V_{S2}, (d) V_{S3}@FH, (e) V_{S3}@DE and (f) V_{S3}@FE systems calculated using the PBE functional. Zero energy is set to the Fermi level.

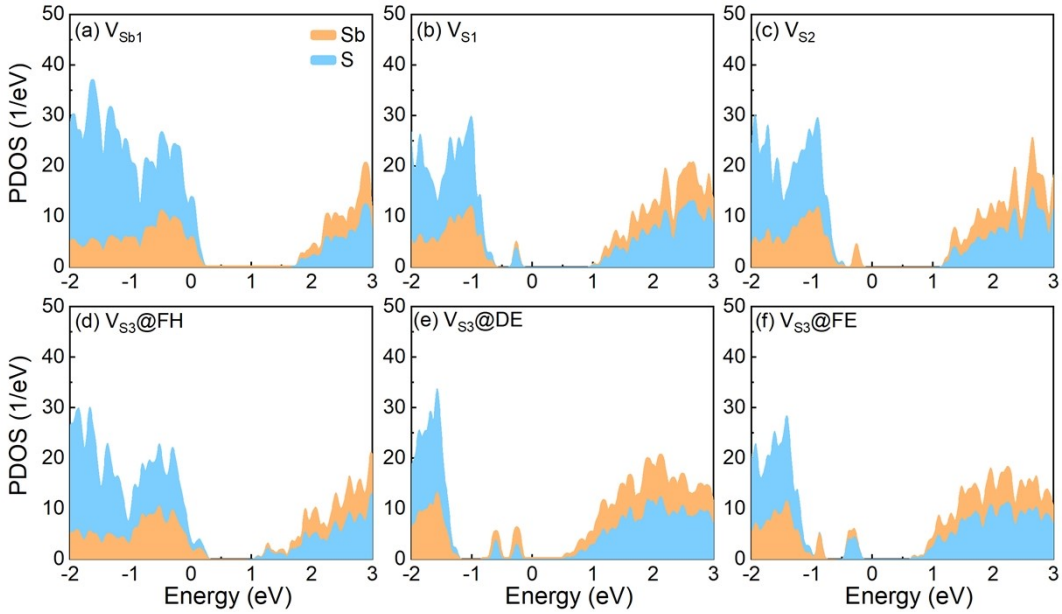

Fig. S9. Projected density of states (PDOS) of (a) V_{Sb1}, (b) V_{S1}, (c) V_{S2}, (d) V_{S3}@FH, (e) V_{S3}@DE and (f) V_{S3}@FE systems calculated using the HSE06 functional. Zero energy is set to the Fermi level.

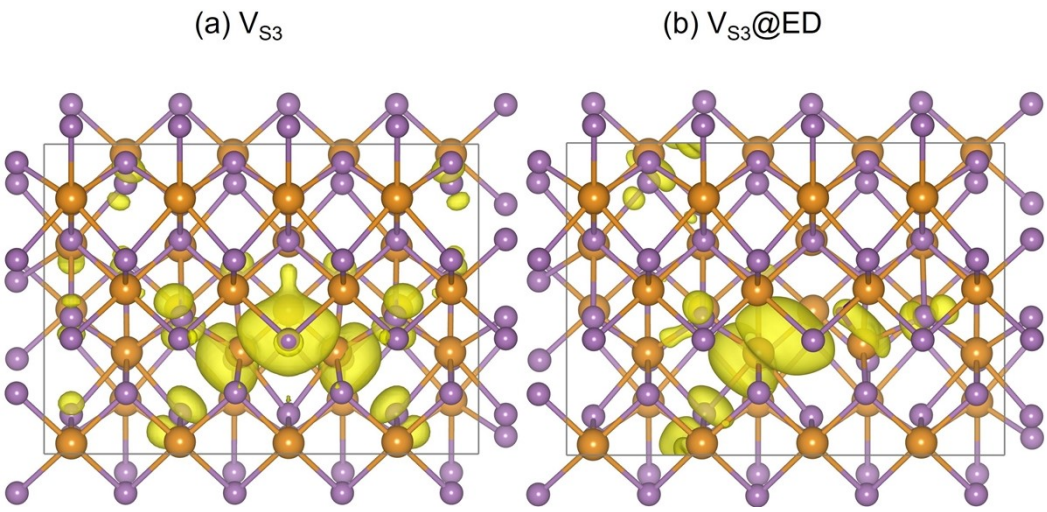

Fig. S10. Charge densities of the high charge density trap state in (a) V_{S3}, and (b) V_{S3}@ED systems.

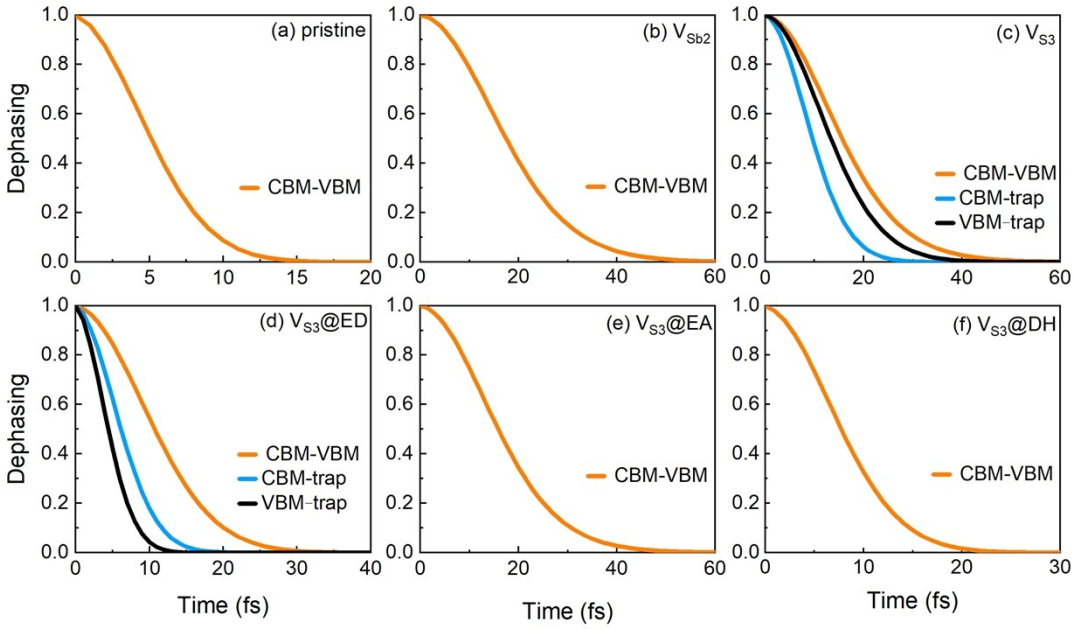

Fig. S11. Pure-dephasing functions for the key pairs of states for charge trapping and recombination in (a) pristine Sb_2S_3, (b) V_{Sb2}, (c) V_{S3}, (d) V_{S3}@ED, (e) V_{S3}@EA and (f) V_{S3}@DH systems.