Supplementary Information (SI) for

Group-IIIA element doped $BaSnS_2$ as a high efficiency absorber for intermediate band solar cell from a first-principles insight

Yang Xue,[a] Changqing Lin,[a] Jiancheng Zhong,[a] Dan Huang[a, b, c*] and Clas Persson[d,e*]

[a] Guangxi Novel Battery Materials Research Center of Engineering Technology, State Key Laboratory of Featured Metal Materials and Life-cycle Safety for Composite Structures, School of Physical Science and Technology, Guangxi University, Nanning 530004, China.

[b] Guangxi Key Laboratory of Precision Navigation Technology and Application, Guilin University of Electronic Technology, Guilin 541004, China.

[c] Guangxi Key Laboratory of Electrochemical Energy Materials, School of Chemistry and Chemical Engineering, Guangxi University, Nanning 530004, China.

[d] Department of Physics and Centre for Materials Science and Nanotechnology, University of Oslo, P.O. Box 1048 Blindern, NO-0316, Oslo, Norway.

[e] Department of Materials Science and Engineering, KTH Royal Institute of Technology, SE-100 44, Stockholm, Sweden.

Corresponding Authors:

*E-mail danhuang@gxu.edu.cn. (Dan Huang), clas.persson@fys.uio.no (Clas Persson)

The calculation detail for the decomposition energy:

In order to investigate their thermal stabilities, the decomposition energies of $BaSnS_2$ with group-IIIA element doping at Sn site at 25% doping concentration were calculated. The decomposition energies (ΔE_D) are calculated by the total energy of $BaSn_{0.75}$IIIA $_{0.25}S_2$(IIIA = Al, Ga and In) subtracting the sum of the total energies of the decomposition products.

Six different decomposition pathways are considered to calculate the decomposition energy:

Path-I:

$$BaSn_{0.75}Al_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{8}BaAl_2S_4 + \frac{1}{8}Ba \quad (\Delta E_D = -0.70\ \text{eV})$$

$$BaSn_{0.75}Ga_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{8}BaGa_2S_4 + \frac{1}{8}Ba$$ (ΔE_D=-0.82 eV)

$$BaSn_{0.75}In_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{8}BaIn_2S_4 + \frac{1}{8}Ba$$ (ΔE_D=-0.92 eV)

Path-II:

$$BaSn_{0.75}Al_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{8}Al_2S_3 + \frac{1}{8}BaS + \frac{1}{8}Ba$$ (ΔE_D=-0.63 eV)

$$BaSn_{0.75}Ga_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{8}Ga_2S_3 + \frac{1}{8}BaS + \frac{1}{8}Ba$$ (ΔE_D=-0.76 eV)

$$BaSn_{0.75}In_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{8}In_2S_3 + \frac{1}{8}BaS + \frac{1}{8}Ba$$ (ΔE_D=-0.83 eV)

Path-III:

$$BaSn_{0.75}Al_{0.25}S_2 \rightarrow \frac{7}{8}BaS + \frac{1}{8}Al_2S_3 + \frac{3}{4}SnS + \frac{1}{8}Ba$$ (ΔE_D=-0.76 eV)

$$8BaSn_{0.75}Ga_{0.25}S_2 \rightarrow \frac{7}{8}BaS + \frac{1}{8}Ga_2S_3 + \frac{3}{4}SnS_2 + \frac{1}{8}Ba$$ (ΔE_D=-1.04 eV)

$$8BaSn_{0.75}In_{0.25}S_2 \rightarrow \frac{7}{8}BaS + \frac{1}{8}In_2S_3 + \frac{3}{4}SnS_2 + \frac{1}{8}Ba$$ (ΔE_D=-0.81 eV)

Path-IV:

$$BaSn_{0.75}Al_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{12}Al_2S_3 + \frac{1}{4}BaS + \frac{1}{6}Al$$ (ΔE_D=-1.54 eV)

$$BaSn_{0.75}Ga_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{12}Ga_2S_3 + \frac{1}{4}BaS + \frac{1}{6}Ga$$ (ΔE_D=-2.09 eV)

$$BaSn_{0.75}In_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{12}In_2S_3 + \frac{1}{4}BaS + \frac{1}{6}In$$ (ΔE_D=-1.82 eV)

Path-V

$$BaSn_{0.75}Al_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{16}BaAl_4S_7 + \frac{1}{8}BaS + \frac{1}{8}Ba$$ (ΔE_D=-0.66 eV)

$$BaSn_{0.75}Ga_{0.25}S_2 \rightarrow \frac{3}{4}BaSnS_2 + \frac{1}{16}BaGa_4S_7 + \frac{1}{8}BaS + \frac{1}{8}Ba \quad (\Delta E_D = -0.79\text{eV})$$

Lack since no crystal structure information of $BaIn_4S_7$ is found.

Path-VI

Lack since no crystal structure information of $Ba_2Al_2S_5$ is found.

$$BaSn_{0.75}In_{0.25}S_2 \rightarrow \frac{5}{8}BaSnS_2 + \frac{1}{8}Ba_2In_2S_5 + \frac{1}{8}SnS + \frac{1}{8}Ba \quad (\Delta E_D = -1.11\ \text{eV})$$

$$BaSn_{0.75}Ga_{0.25}S_2 \rightarrow \frac{5}{8}BaSnS_2 + \frac{1}{8}Ba_2Ga_2S_5 + \frac{1}{8}SnS + \frac{1}{8}Ba \quad (\Delta E_D = -1.01\ \text{eV})$$

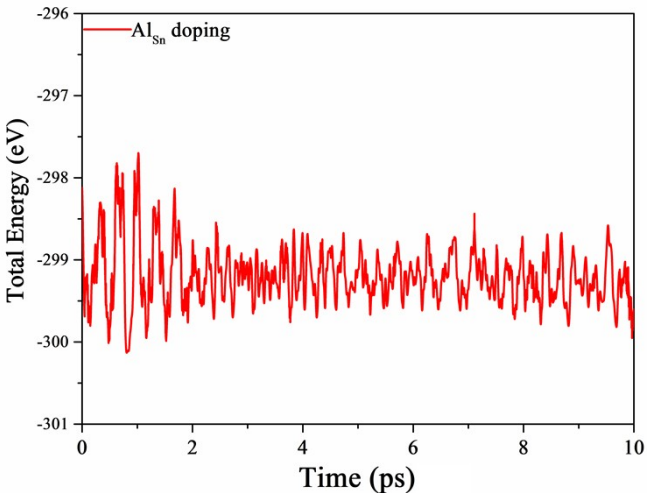

Fig. S1. Fluctuation of the free energy of $BaSnS_2$ with Al doping at Sn site at 25% doping concentration during the Ab initio molecular dynamic simulation at 300 K within 10 ps.

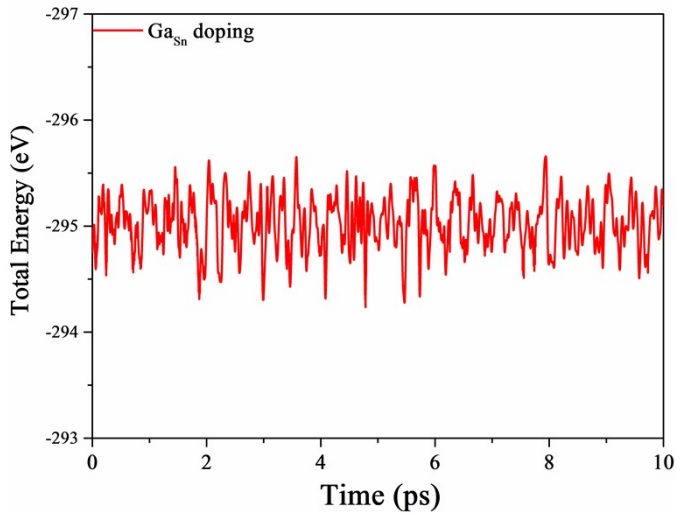

Fig. S2. Fluctuation of the free energy of $BaSnS_2$ with Ga doping at Sn site at 25% doping concentration during the Ab initio molecular dynamic simulation at 300 K within 10 ps.

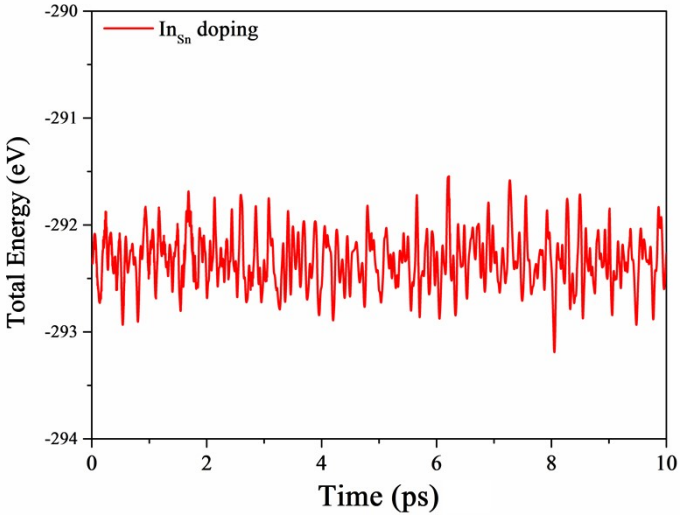

Fig. S3. Fluctuation of the free energy of $BaSnS_2$ with In doping at Sn site at 25% doping concentration during the Ab initio molecular dynamic simulation at 300 K within 10 ps.

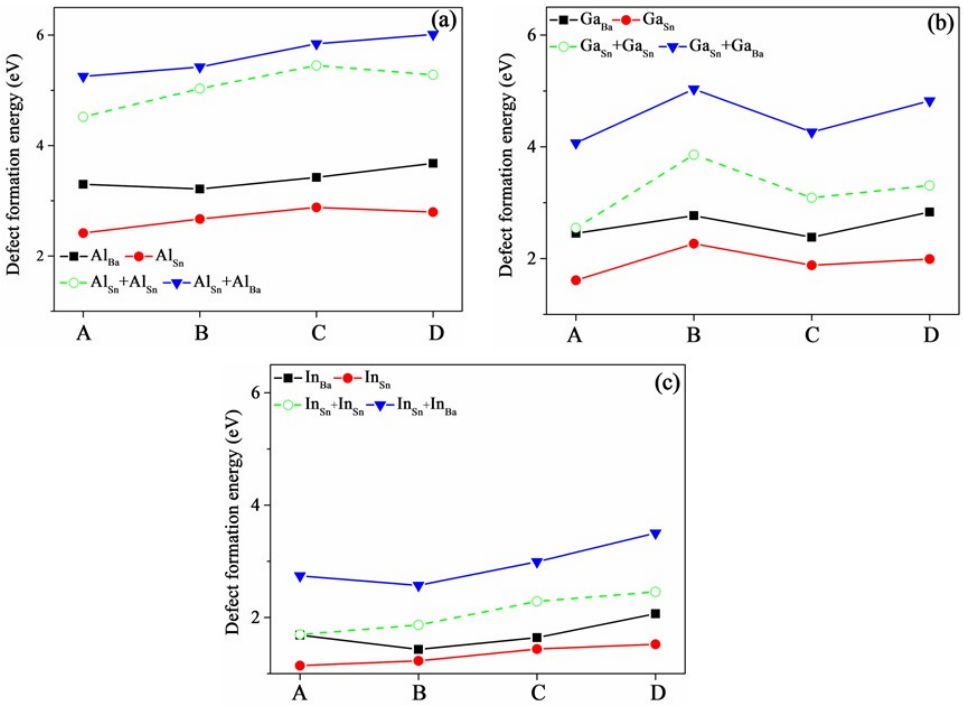

Fig. S4. The formation energies of substitution defects and their complexes in group-IIIA element doped $BaSnS_2$ by using 2×1×1 supercells.

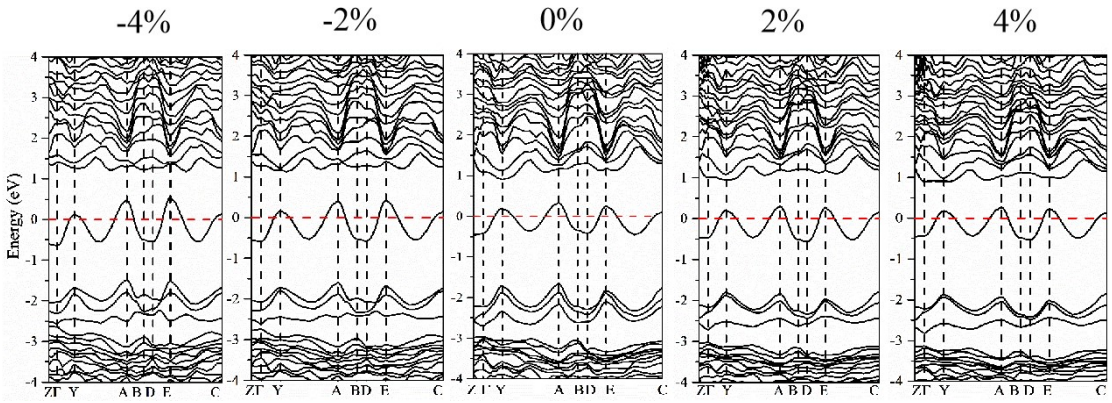

Fig. S5. The band structures of $BaSnS_2$ with Al doping at Sn site after the implementation of the strain in the range of -4%-4%.

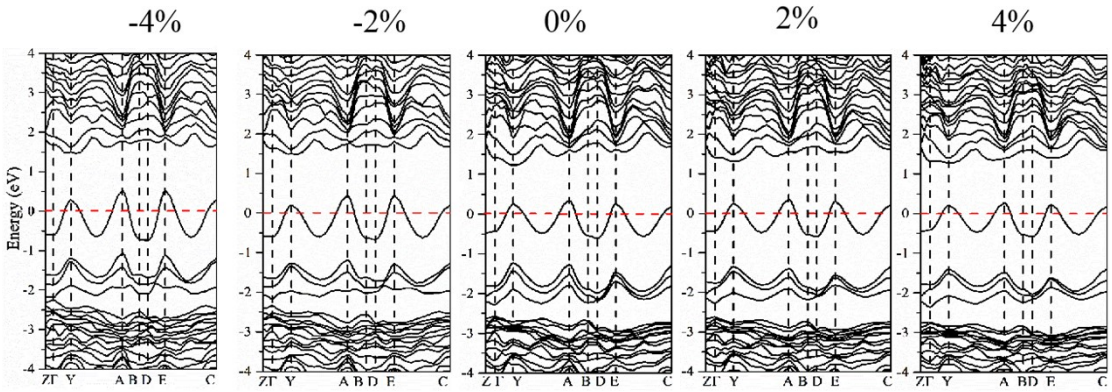

Fig. S6. The band structures of $BaSnS_2$ with Ga doping at Sn site after the implementation of the strain in the range of -4%-4%.

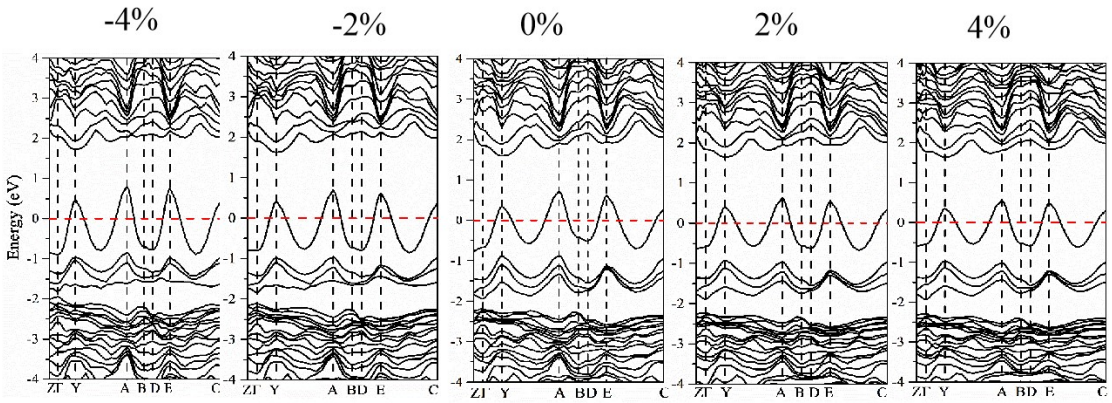

Fig. S7. The band structures of $BaSnS_2$ with In doping at Sn site after the implementation of the strain in the range of -4%-4%.

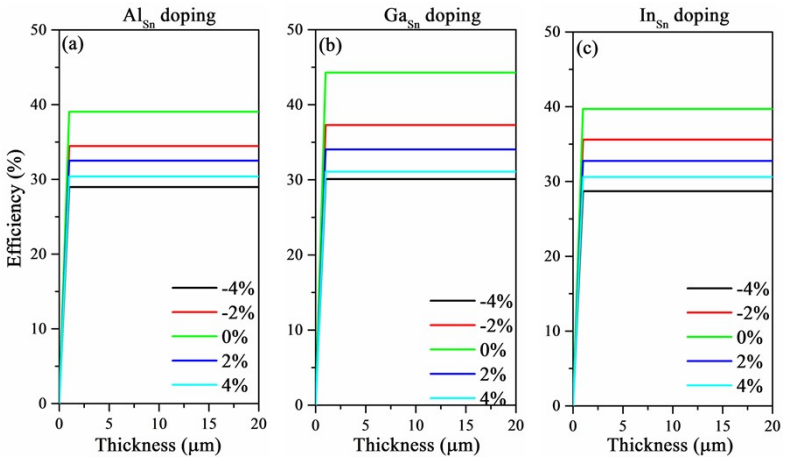

Fig. S8. The theoretical efficiencies of $BaSnS_2$ with group-IIIA element doping at Sn site at 25% doping concentration as a function of absorber thickness after considering the strain engineering in the range of -4%-4%.

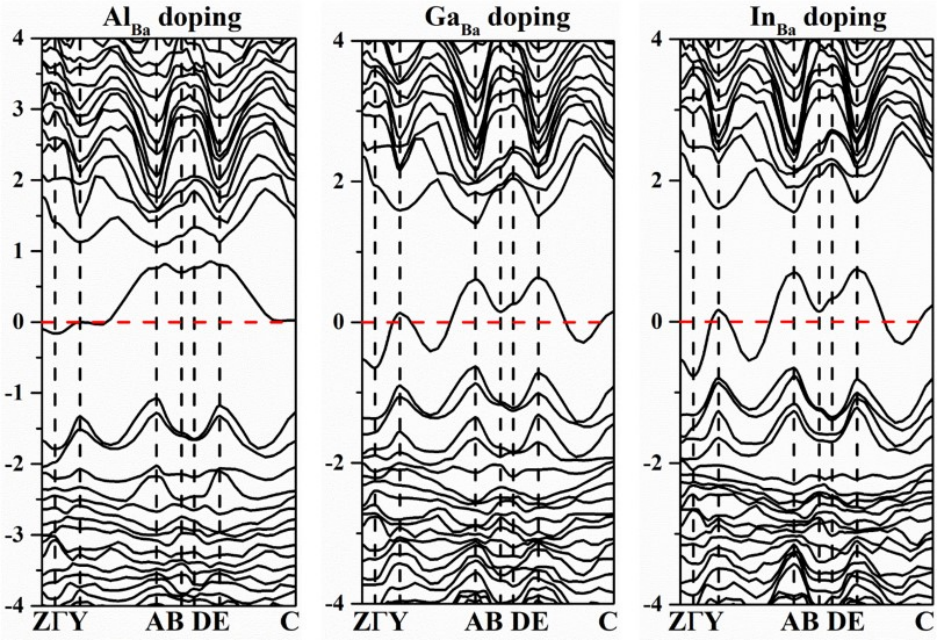

Fig. S9. The band structures of $BaSnS_2$ with group-IIIA element doping at Ba site at 25% doping concentration.