

Supporting information

p-type Co interstitial defects in thermoelectric skutterudite CoSb_3 due to the breakage of Sb_4 -rings

Guodong Li ^{†‡}, Saurabh Bajaj [§], Umut Aydemir [‡], Shiqiang Hao [‡], Hai Xiao [⊥], William A. Goddard III ^{*⊥}, Pengcheng Zhai [†], Qingjie Zhang ^{*†}, and G. Jeffrey Snyder ^{*‡}

[†]State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan, Hubei 430070, China.

[‡]Department of Materials Science and Engineering, Northwestern University, Evanston, Illinois 60208, United States.

[§]Environmental Energy & Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States.

[⊥]Materials and Process Simulation Center, California Institute of Technology, Pasadena, California 91125, United States.

*Corresponding authors: jeff.snyder@northwestern.edu; wag@wag.caltech.edu; zhangqj@whut.edu.cn

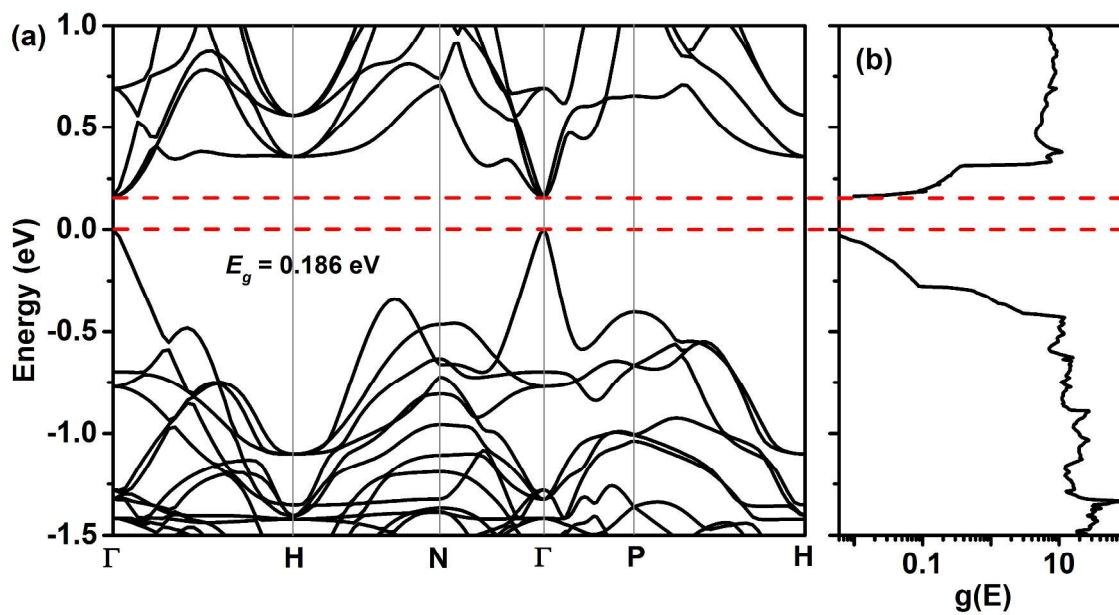


Figure S1. (a) The band structure and (b) density of state of CoSb_3 from our *ab-initio* calculation using PBE functional. The direct band gap $E_g = 0.186$ eV.

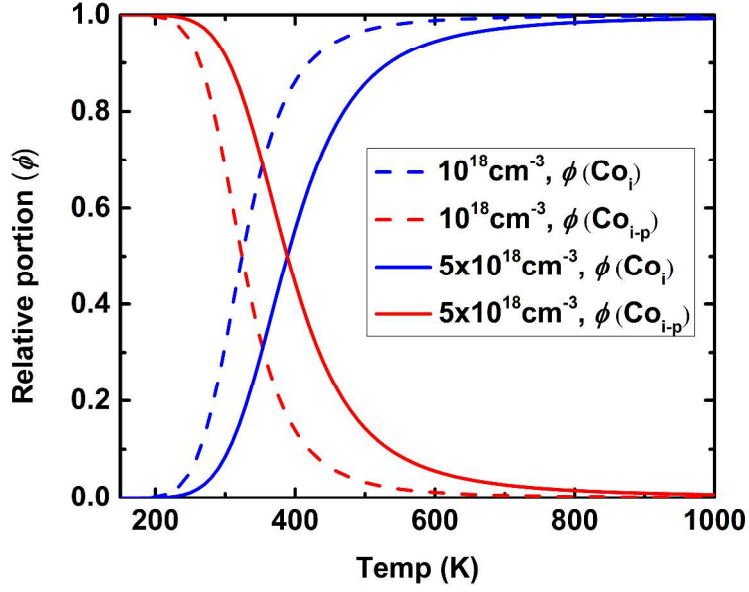


Figure S2. Relative portion of Co_i and Co_{i-p} as a function of the temperature. Solid line and dot line represent 5 × 10¹⁸ cm⁻³ and 10¹⁸ cm⁻³ carriers in the system respectively. The relation portion is calculated by

$$\phi = \frac{1}{1 + \exp\left(\frac{\Delta H - T \Delta S}{kT}\right)},$$

where ΔH is the formation enthalpy change between neutral Co_i and Co_{i-p}.

ΔS is the configurational entropy change defined by equation (3) in the manuscript. The defect thermodynamics calculation is time-independent.