

# Supporting Information for “Highly-Robust Reentrant Superconductivity in CsV<sub>3</sub>Sb<sub>5</sub> under Pressure”

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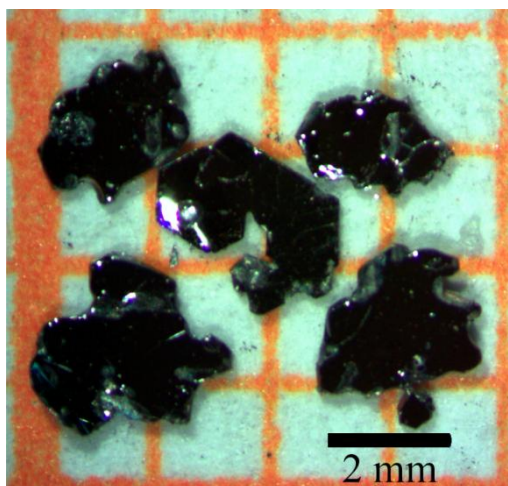
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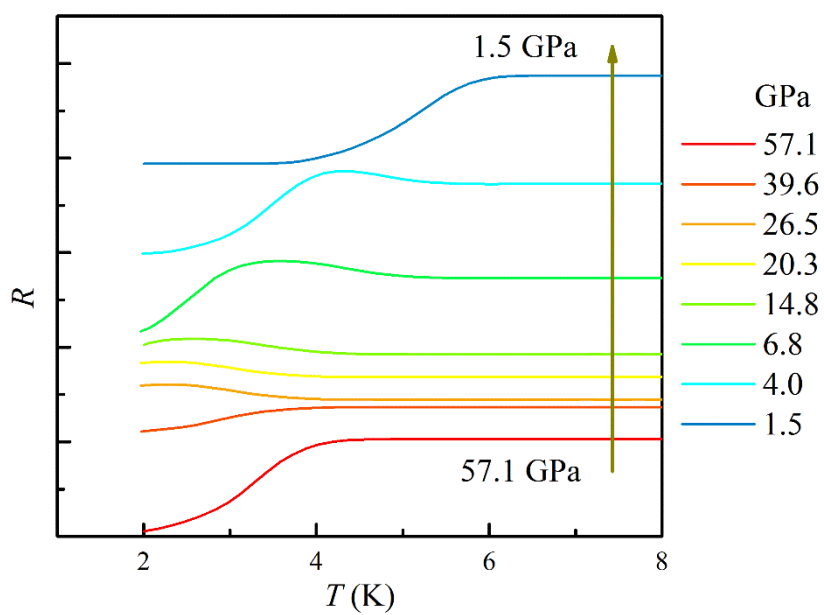
Email: [xiaobing.phy@qfnu.edu.cn](mailto:xiaobing.phy@qfnu.edu.cn); [jgguo@iphy.ac.cn](mailto:jgguo@iphy.ac.cn); [chenx29@iphy.ac.cn](mailto:chenx29@iphy.ac.cn)

**Table S1.** Change of crystallographic parameters for CsV<sub>3</sub>Sb<sub>5</sub> with respect to pressure in the range of 0-100 GPa.

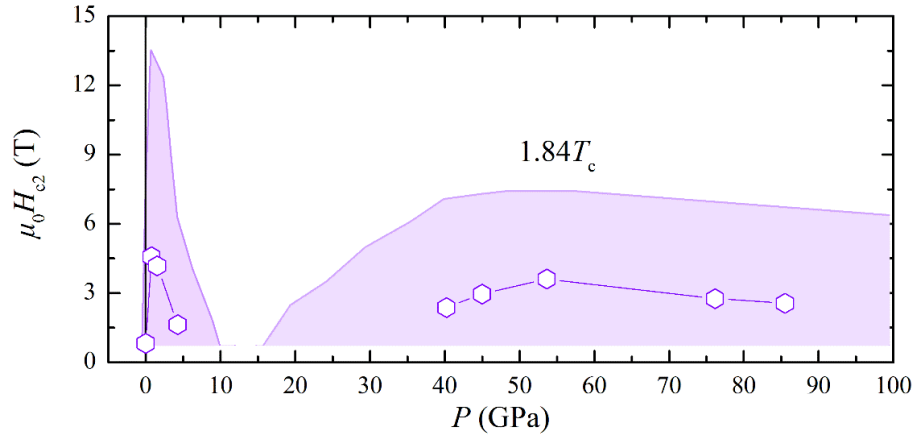
Phase	Pressure (GPa)	Lattice parameters (Å, °)	Wuckoff position (fractional)			
			Atoms	x	y	z
<i>P6/mmm</i>	0	a = b = 5.49220	Cs(1a)	0.0000	0.0000	0.0000
		c = 9.88870	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.7292
<i>P6/mmm</i>	5	a = b = 5.42020	Cs(1a)	0.0000	0.0000	0.0000
		c = 8.58070	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.7612
<i>P6/mmm</i>	10	a = b = 5.3598	Cs(1a)	0.0000	0.0000	0.0000
		c = 8.11750	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.7734
<i>P6/mmm</i>	20	a = b = 5.25530	Cs(1a)	0.0000	0.0000	0.0000
		c = 7.68210	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.7840
<i>P6/mmm</i>	40	a = b = 5.10680	Cs(1a)	0.0000	0.0000	0.0000
		c = 7.19870	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.7940
<i>P6/mmm</i>	60	a = b = 5.01490	Cs(1a)	0.0000	0.0000	0.0000
		c = 6.84900	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.8000
<i>P6/mmm</i>	100	a = b = 4.85330	Cs(1a)	0.0000	0.0000	0.0000
		c = 6.52460	V(3g)	0.5000	0.5000	0.5000
		α = γ = 90.00	Sb(1b)	0.0000	0.0000	0.5000
		β = 120.00	Sb(4h)	0.6667	0.3333	0.8041



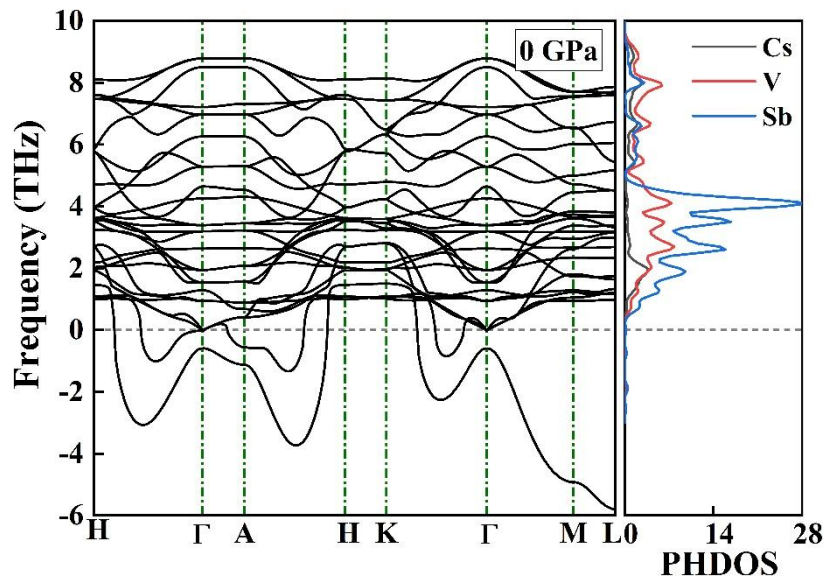
**Figure S1.** Optical photograph of CsV<sub>3</sub>Sb<sub>5</sub> single crystals.



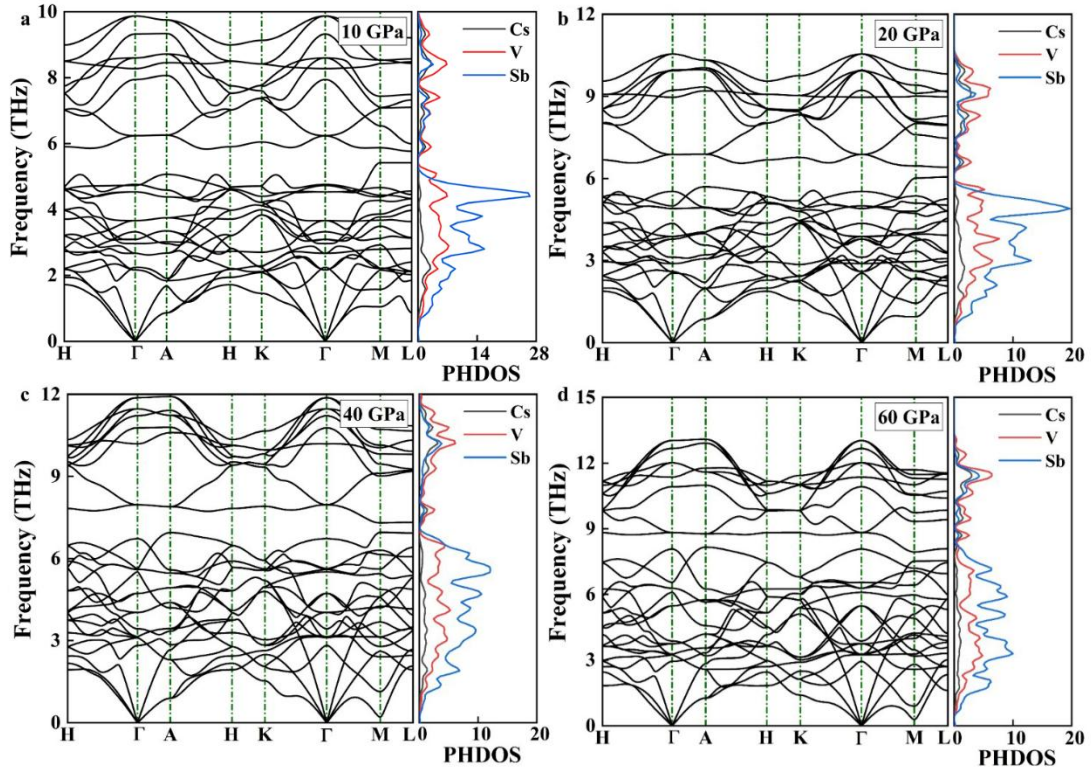
**Figure S2.** Temperature-dependent resistance of CsV<sub>3</sub>Sb<sub>5</sub> at various pressures of 57.1-1.5 GPa,



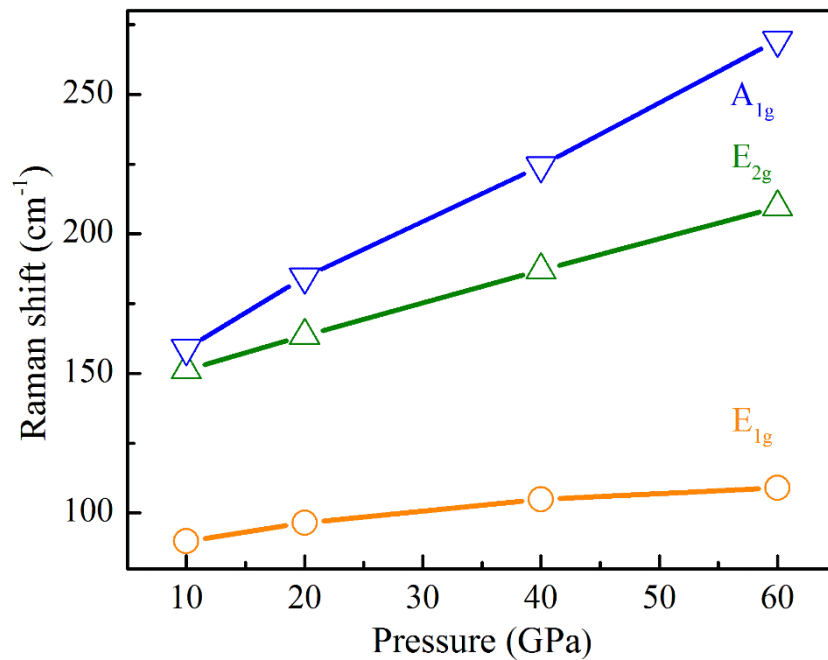
**Figure S3.** Pressure dependences of the upper critical field  $\mu_0 H_{c2}(0)$  obtained from the linear fitting.



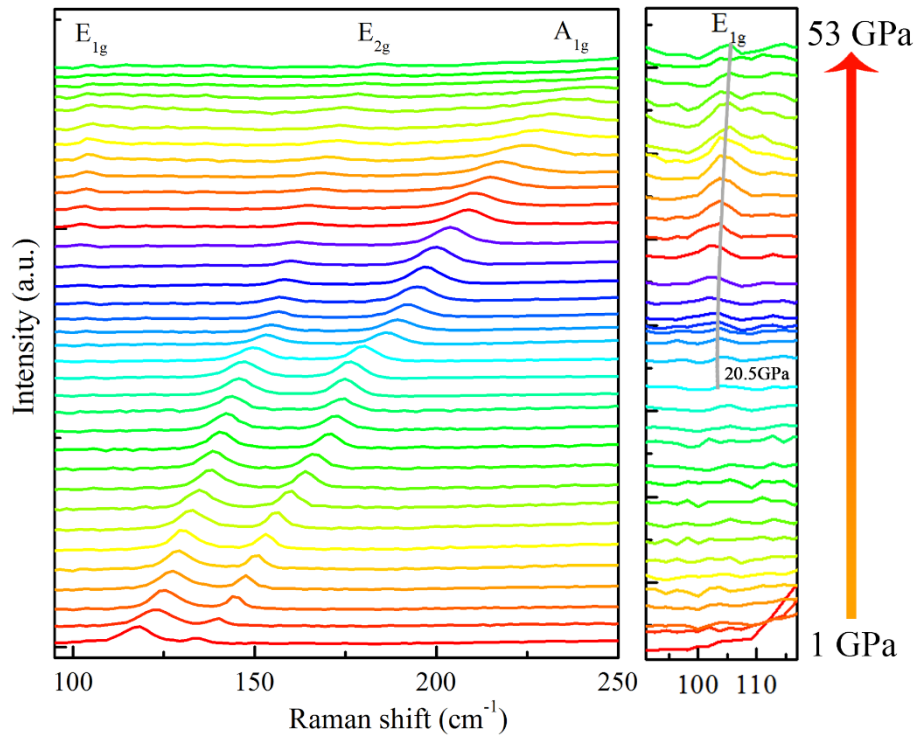
**Figure S4.** Calculated phonon dispersions and phonon density of states (PHDOS) of  $\text{CsV}_3\text{Sb}_5$  at 0 GPa.



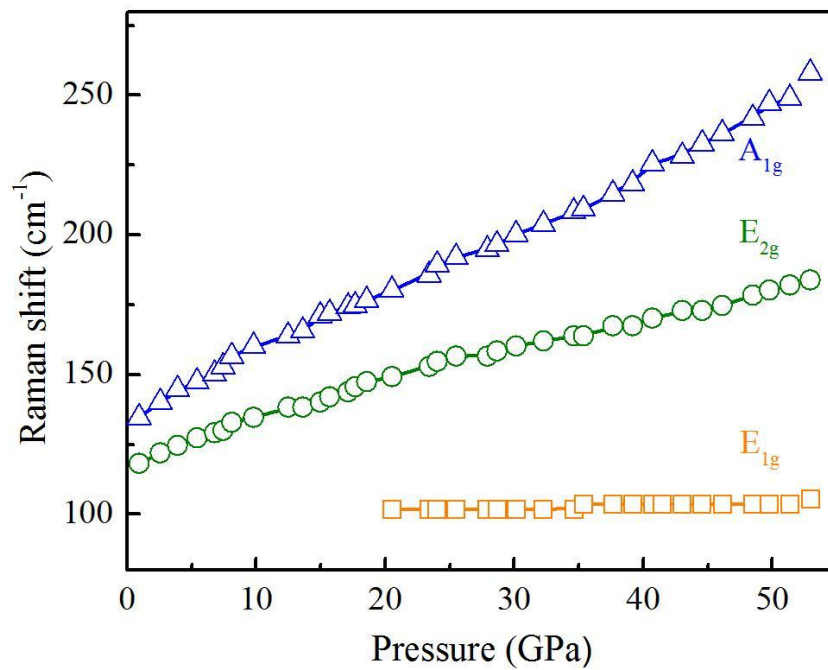
**Figure S5.** Calculated phonon dispersions and phonon density of states (PHDOS) of  $\text{CsV}_3\text{Sb}_5$  at (a) 10 GPa, (b) 20 GPa, (c) 40 GPa, and (d) 60 GPa, respectively.



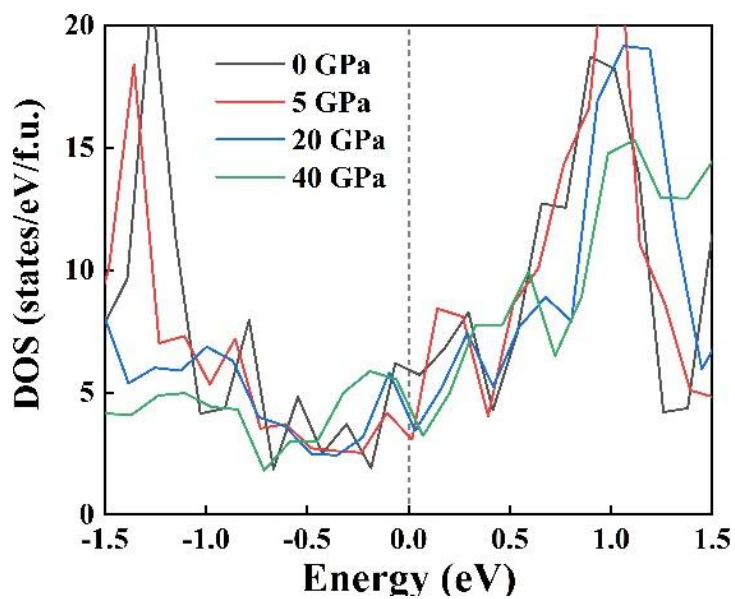
**Figure S6.** Pressure versus Raman frequency of  $\text{CsV}_3\text{Sb}_5$  from phonon dispersions at 10 GPa, 20 GPa, 40 GPa and 60 GPa, respectively.



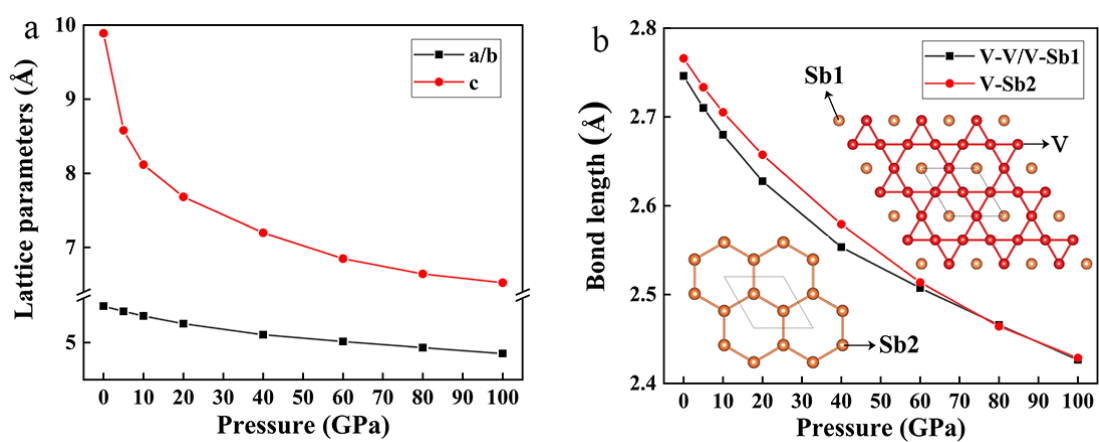
**Figure S7.** Pressure-induced Raman changes of  $\text{CsV}_3\text{Sb}_5$  in the range of 1-53 GPa.



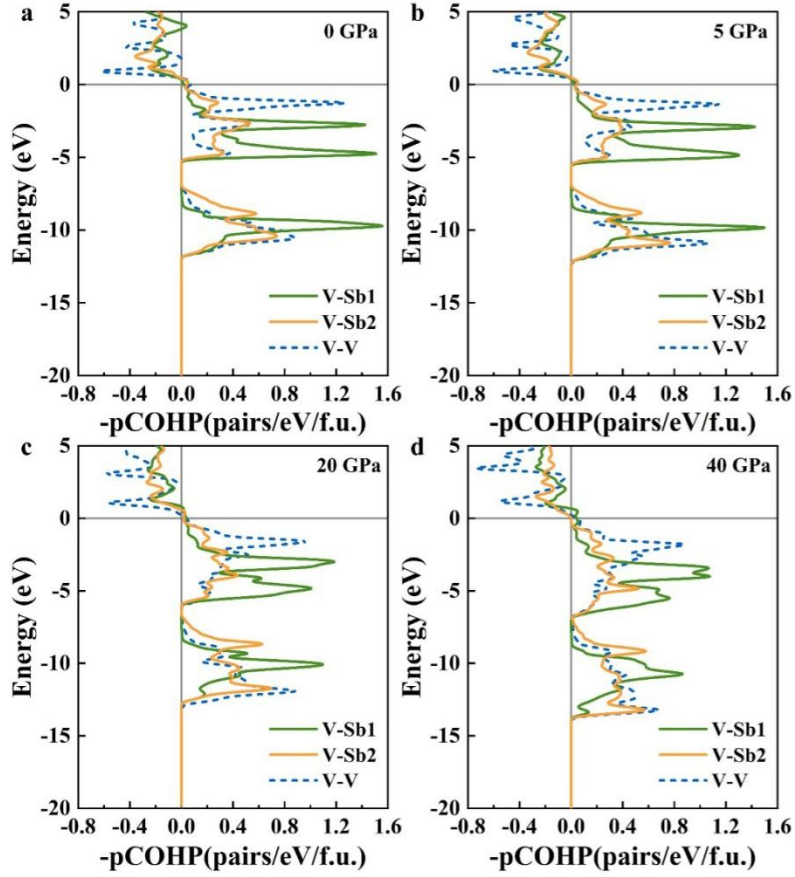
**Figure S8.** Pressure versus Raman frequency of  $\text{CsV}_3\text{Sb}_5$  in the range of 1-53 GPa.



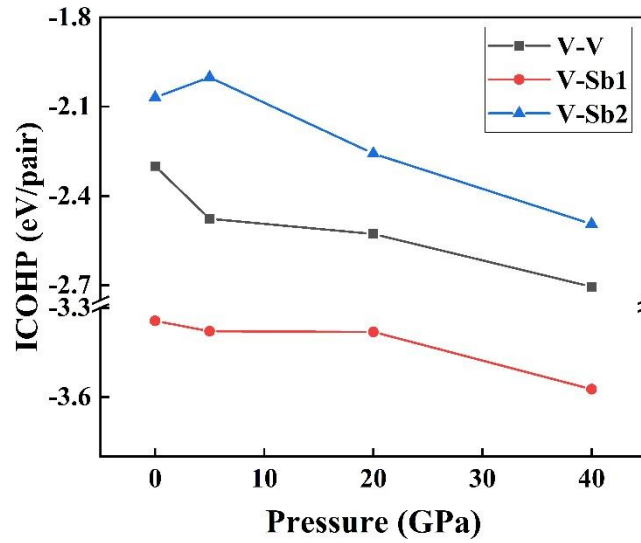
**Figure S9.** Calculated total density of states (DOS) for  $\text{CsV}_3\text{Sb}_5$  at selected pressures.



**Figure S10.** The change of lattice parameters (a) and bond lengths (b) with respect to pressure in the range of 0-100 GPa.



**Figure S11.** Projected crystal orbital Hamiltonian Population (-pCOHP) of CsV<sub>3</sub>Sb<sub>5</sub> at (a) 0 GPa, (b) 5 GPa, (c) 20 GPa and (d) 40 GPa, respectively. The values of -pCOHP >0 signify bonding states and the values of -pCOHP <0 signify antibonding states. The Fermi level is set to zero.



**Figure S12.** The integrated COHP (ICOHP) for V-V, V-Sb1 and V-Sb2 bonds of CsV<sub>3</sub>Sb<sub>5</sub> at the range from 0 to 40 GPa.