Supporting Information for “Highly-Robust Reentrant Superconductivity in CsV$_3$Sb$_5$ under Pressure”

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Table S1. Change of crystallographic parameters for CsV₃Sb₅ with respect to pressure in the range of 0-100 GPa.

| Phase | Pressure (GPa) | Lattice parameters (Å, °) | Wuckoff position (fractional) |
|-------|---------------|---------------------------|-----------------------------|
|       |               |                           | Atoms | x     | y     | z     |
| P6/mmm| 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 |
|       |               | 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 |
|       |               | 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 |
|       |               | 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 |
|       |               | 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 |
|       |               | 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 |
|       |               | 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$_3$Sb$_5$ single crystals.

Figure S2. Temperature-dependent resistance of CsV$_3$Sb$_5$ 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 CsV$_3$Sb$_5$ at 0 GPa.
Figure S5. Calculated phonon dispersions and phonon density of states (PHDOS) of CsV$_3$Sb$_5$ at (a) 10 GPa, (b) 20 GPa, (c) 40 GPa, and (d) 60 GPa, respectively.

Figure S6. Pressure versus Raman frequency of CsV$_3$Sb$_5$ from phonon dispersions at 10 GPa, 20 GPa, 40 GPa and 60 GPa, respectively.
Figure S7. Pressure-induced Raman changes of CsV$_3$Sb$_5$ in the range of 1-53 GPa.

Figure S8. Pressure versus Raman frequency of CsV$_3$Sb$_5$ in the range of 1-53 GPa.
Figure S9. Calculated total density of states (DOS) for CsV$_3$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 CsV₃Sb₅ 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 CsV₃Sb₅ at the range from 0 to 40 GPa.