

1 **Supplemental Materials**

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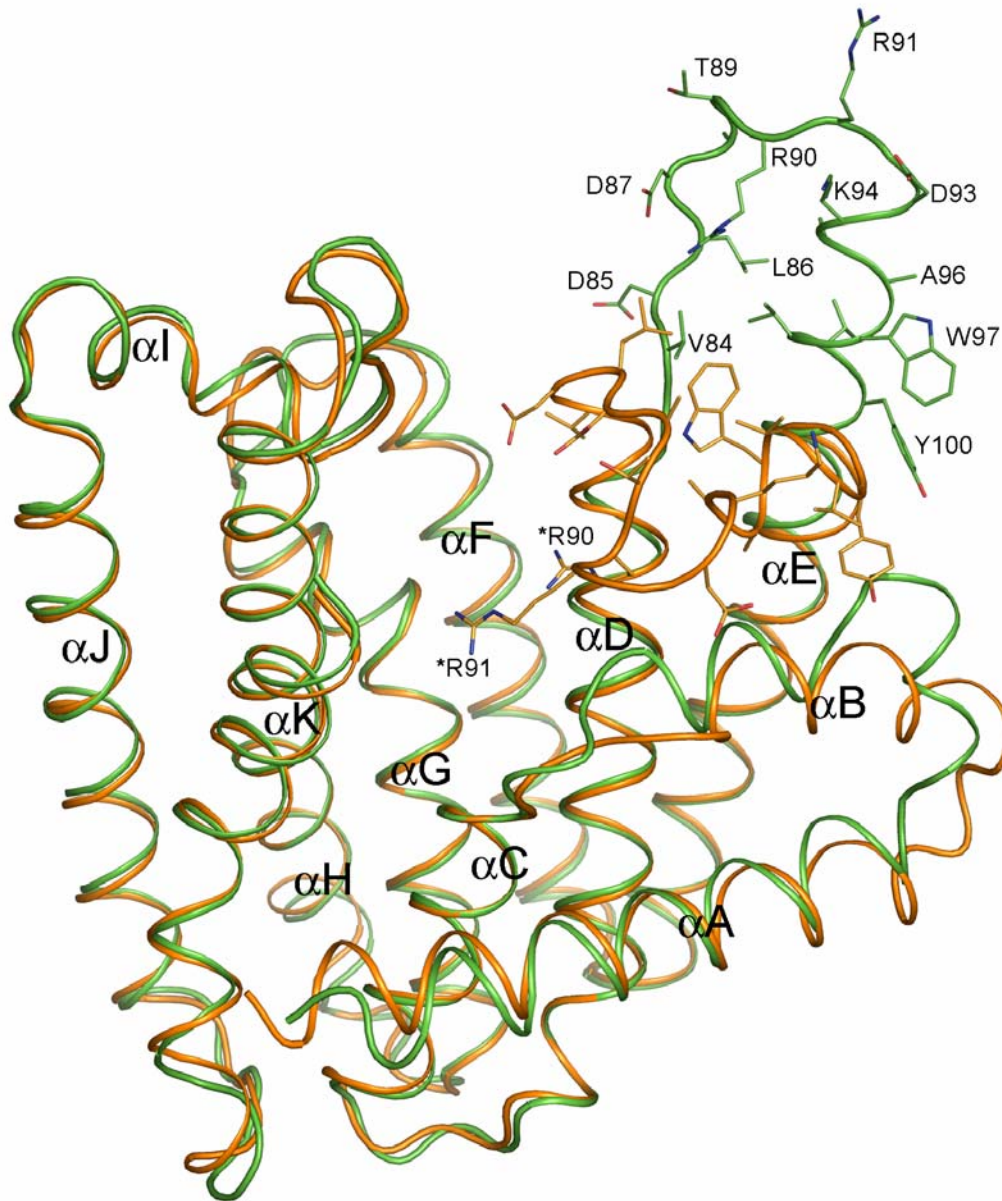
3 **TABLE S1. Primers used to construct HexPPs mutants in this study.**

Mutant	Primer sequence
D81C	5'-CCATTCTGCTTCCTTAGCTTTGTGTGATATAGTTGATCTCGACG-3'
Y124A	5'-GCTCTAAGGATAATTCAGACTTCTGCTGGTGACGATGCTTTA-3'
Y174A	5'-TCTACGGTACTATCAGCCGCCGCTTCTAAACACTATAATACG-3'
L164A	5'-TTAAAAACCGGTAGTGCATTTAAGCTCTCTACGGTACTATCAGCC-3'
L164G	5'-TTAAAAACCGGTAGTGGATTTAAGCTCTCTACGGTACTATCAGCC-3'
W136E	5'-TACTAGTATTGAGTTAGAGAAAGACACCTCAGTGGGTGCTCTAAG-3'
F117E^a	5'-GATCTTGTACTTGTTCAGTGCGGAACATACGGTTGAAGAGATAGGA-3'

4 ^a F117E OPPs mutant of *T. maritima*

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1 Figure S1. The two subunits of *S. solfataricus* HexPPs are superimposed. Monomer A
2 and B are shown in orange and green, respectively. The residues 84-100 shown in
3 stick model cannot superimposed. The diagram was produced with PyMOL.

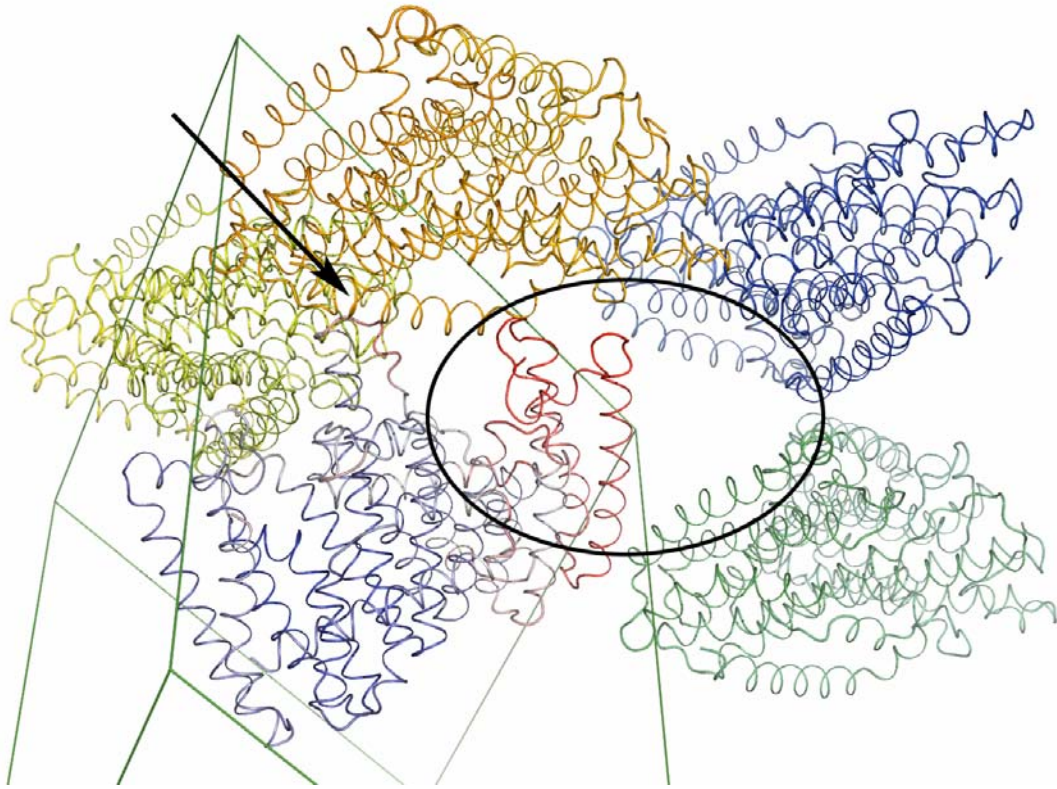


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Sun *et al.*, Figure S1

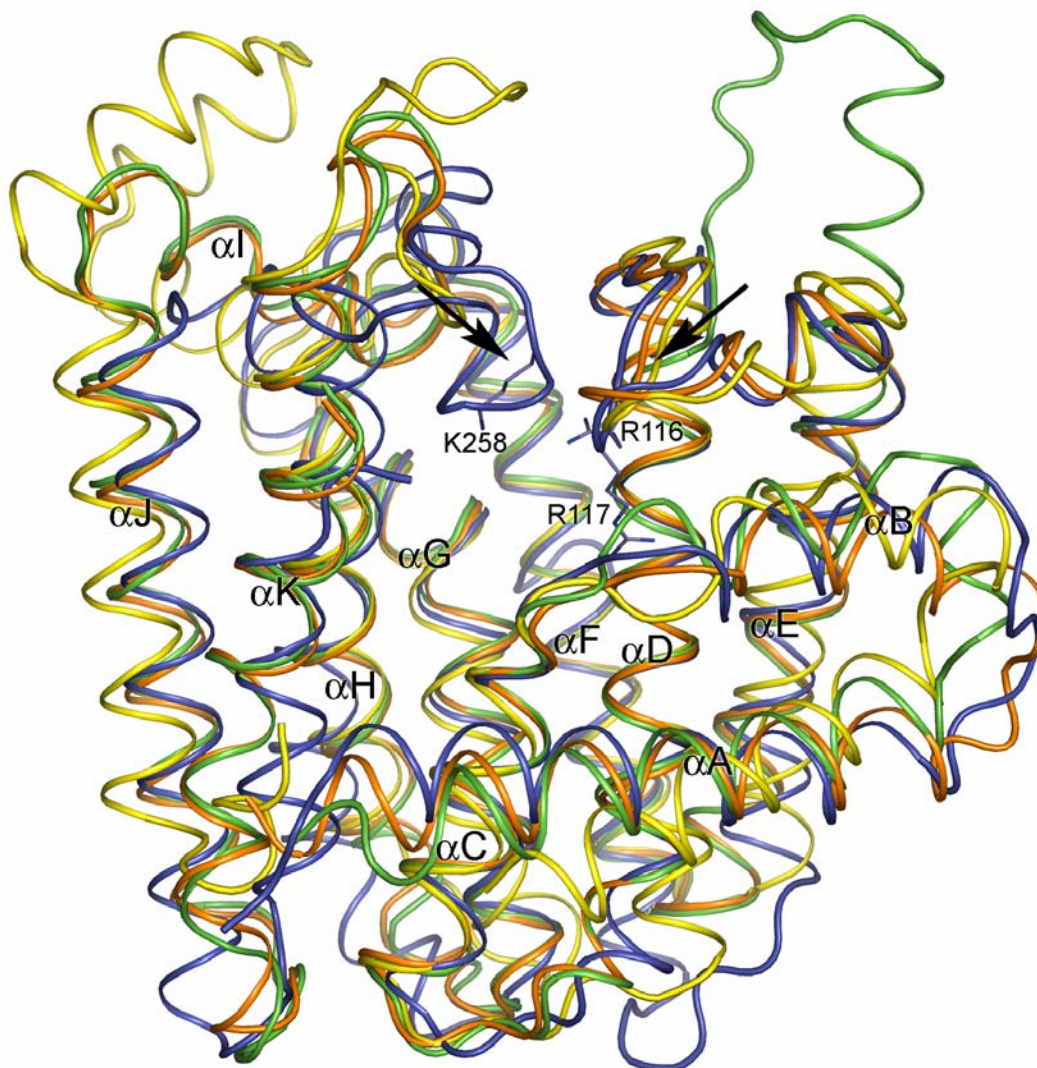
1 Figure S2. The neighboring HexPPs molecules in the symmetry-related unit cells
2 are shown in different colors. The arrow indicates the helices D and E of
3 monomer B interacting with the neighboring HexPPs enzymes. The oval indicates
4 that there is space for helices H-K in monomer B to move, and this may cause
5 high B factors of monomer B. The diagram was produced with PyMOL.



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Sun *et al.*, Figure S2

1 Figure S3. The three *trans*-PTases are superimposed. The FPPs complex of Protein
2 Data Bank structure 1RQI is shown in blue, the OPPs of 1V4E is shown in yellow,
3 and the monomer A and B of HexPPs crystal structures studied here are in orange and
4 green, respectively. Monomer A of HexPPs has a similar conformation to OPPs of
5 1V4E. FPPs complex reveals active-site conformational changes localized at the
6 α D- α E and α H- α J loops (arrow) (10). The residues R116 and R117 in α D- α E loop,
7 residue K258 in α H- α J loop are shown in stick. The diagram was produced with
8 PyMOL.



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Sun *et al.*, Figure S3