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Direct Observation of Thymine Dimer Repair in DNA by Photolyase

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Departments of Physics, Chemistry, and Biochemistry, Programs of Biophysics, Chemical Physics, and Biochemistry, The Ohio State University, Columbus, 191 West Woodruff Avenue, OH 43210. Photolyase uses light energy to split ultraviolet-induced cyclobutane pyrimidine dimers in damaged DNA, but its molecular mechanism has never been directly revealed. We report here the direct mapping of catalytic processes through femtosecond synchronization of the enzymatic dynamics with the repair function. We observed direct electron transfer from the excited flavin cofactor to the dimer in 170 ps and back electron transfer from the repaired thymines in 560 ps. Both reactions are strongly modulated by active-site solvation to achieve maximum repair efficiency. These results show that the photocycle of DNA repair by photolyase is through a radical mechanism and completed on subnanosecond time scale at the dynamic active site with no net electron change in redox states of the flavin cofactor.