Photolyase splits cyclobutane ring of pyrimidine dimer (CPD) in DNA in a light (350-500 nm) driven reaction and thus reverses the harmful effects of far-UV (200-300 nm). Through resonance energy transfer from photoantenna MTHF or directly by absorption of visible light, reduced flavin cofactor (FADH\(^{-}\)) transfers an electron to CPD which subsequently splits into two pyrimidines. Concomitantly, an electron is transferred back to the oxidized neutral radical (FADH\(^{+}\)) to restore the active form (FADH\(^{+}\)). With femtosecond resolution we followed the functional evolution and observed forward and backward electron transfer reactions as well as the entire DNA-repair process. These dynamics occur in the picosecond time scale, unraveling the ultrafast nature of DNA-repair mechanism.