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EXPLORING STRUCTURAL DYNAMICS WITH TIME-RESOLVED DIFFRACTION

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Through the advancement of computational methods, prediction of transient molecular structures has become relatively easy. In addition, molecular dynamics is now routinely studied on femtosecond time scales using various spectroscopies. However, typical spectroscopic methods cannot provide direct structural information of all nuclear coordinates involved in such dynamical processes, and direct experimental verification of such structures of short lifetime is difficult. Currently, the only available method that provides both direct structural information of atomic scale and ultrafast time resolution of sub-picosecond and picosecond is time-resolved diffraction by either electrons or x-rays. Typical time-resolved diffraction experiments are conducted in a pump-probe scheme; femtosecond laser pulses are used to initiate the reaction in the molecules of interest, and after a well-defined time delays, x-rays or electrons are sent to the sample to probe the transient structures in the course of reaction. Here we report direct structural observation of the long-sought bridged radical in solution using time-resolved liquid-phase x-ray diffraction. The structural dynamics spanning picosecond to microsecond time scale has been experimentally determined in unprecedented structural details.

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