Abstract Submitted
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Tracking picosecond molecular dynamics in solution using a suite of synchrotron-x-ray spectroscopic tools[1] ANNE MARIE MARCH, GILLES DOUMY, ELLIOT P. KANTER, STEFAN LEHMANN, DOOSHAYE MOONSHIRAM, STEPHEN H. SOUTHWORTH, LINDA YOUNG, Argonne National Lab, TADESSE A. ASSEFA, CHRISTIAN BRESSLER, WOJCIECH GAWELDA, European XFEL, ZOLTÁN NÉMETH, GYÖRGY VANKÓ, Hungarian Academy of Sciences — Laser-pump, X-ray-probe techniques are powerful tools for exploring molecular structural changes that occur in complex environments such as solutions, during a photo-initiated reaction. We are developing such methods using hard x-rays from the Advanced Photon Source, combining x-ray emission spectroscopy and x-ray absorption spectroscopy as probes of electronic and geometric structure and using high-power, MHz lasers as pumps. The high-duty-cycle pump-probe measurements efficiently utilize the synchrotron x-ray flux and enable high-fidelity measurements of the structures of transient intermediates. We present measurements on the model system $[Fe(II)(CN)_6]^{4-}$ (ferrocyanide) in an aqueous solution after excitation with 355 nm and 266 nm laser light. The system undergoes two wavelength dependent reactions: photooxidation and photoaquation. Iron K-edge absorption spectra were obtained along with iron emission spectra. Our data support the presence of a previously unobserved pentacoordinated intermediate species in the photoaquation reaction. Its lifetime has been measured to be 4.6 ns and details of its structure will be discussed.

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