

Femtosecond Time-Resolved X-ray Absorption Spectroscopy of Liquids using SACLA

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We present time-resolved X-ray absorption spectroscopy (TRXAS) of aqueous solutions using a hard X-ray free electron laser (SACLA: SPring-8 Angstrom Compact Free Electron Laser). An aqueous solution of $[\text{Fe}(\text{III})(\text{C}_2\text{O}_4)_3]^{3-}$ is photoexcited by a femtosecond 266 nm pulse, and transient X-ray absorption spectra were measured as a function of pump-probe delay time. The Fe K-edge exhibited an instantaneous red-shift upon 266 nm photoexcitation followed by a slight blue shift within 2 ps and remained unchanged over 100 ps. The X-ray fluorescence method offers higher sensitivity than a direct absorption method [Obara et al. Optics Express, 22, 1105 (2014)] and enables TRXAS at one order of magnitude lower sample concentration. Comparison with TDDFT calculations of the K-edges of the product candidates reveals that the most likely products within 100 ps are $[\text{Fe}(\text{II})(\text{C}_2\text{O}_4)_2]^-$ or $[\text{Fe}(\text{III})(\text{C}_2\text{O}_4)_2]^{2-}$.

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