

Abstract Submitted
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Using time-resolved Ru L-edge X-ray absorption spectroscopy to capture photoinduced transient electronic structure in a solar cell dye molecule BENJAMIN VAN KUIKEN, MICHAEL LYNCH, MUNIRA KHALIL, Department of Chemistry, University of Washington, Seattle, WA 98195, NILS HUSE, HANA CHO, MATTHEW STRADER, ROBERT SCHOENLEIN, Lawrence Berkeley National Laboratory, Berkeley, CA 94720 — Understanding the electronic structure of transition metal dye molecules used in dye-sensitized solar cells (DSSC) is critical for determining the functioning of these devices. Ru^{II} dyes such as Ru(dcbpy)₂(NCS)₂ (termed RuN3) have been components in some of the most effective DSSCs. We use synchrotron-based picosecond Ru L-edge X-ray absorption spectroscopy (XAS) to monitor changes the changes in the electronic structure of RuN3⁴⁻ that accompany the ¹A₁ to ³MLCT conversion initiated with a 400 nm light pulse. The results are interpreted by simulating the Ru L₃ X-ray absorption spectra of the ¹A₁ and ³MLCT states with time-dependent density functional theory (TD-DFT). We observe the formation of the Ru^{III} oxidation state of RuN3⁴⁻ within the 70 ps time resolution of our experiment. The TD-DFT simulation allows us to assign a spectral feature in the Ru L-edge spectrum as a probe of the electronic structure of the NCS ligands due to overlap between Ru 4d and NCS π* orbitals. A 1.2 eV blue shift in this feature between ground and ³MLCT state corresponds to depletion in charge density on the NCS ligands in the excited state. We will discuss in detail the local electronic structure around the Ru atom in the transient ³MLCT state measured by time-resolved XAS.

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