

USING POLARIZED ULTRAFAST SPECTROSCOPY AS A TOOL FOR DETERMINING SUBPICOSECOND STRUCTURAL DYNAMICS OF PHOTOEXCITED N-PROPYLCOBALAMIN

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Pump-probe spectroscopy has been utilized for decades to take ultrafast “snapshots” of molecular systems as they traverse their excited state surfaces. Recently, linearly polarized pump and probe pulses have been manipulated to separate “magic angle” X-ray spectra from isotropic solutions into two components: signal from the transition dipole moment direction and signal from the two orthogonal directions. This polarized ultrafast spectroscopy provides more electronic and/or structural information compared to using magic angle. In the study presented here, polarized X-ray absorption near-edge structure (XANES) at the Cobalt K-edge and UV-visible transient absorption (TA) spectroscopy were used to monitor the evolution of the excited-state structure of n-propylcobalamin following visible excitation. Finite difference method near edge structure (FDMNES) is used in conjunction with polarized XANES experimental data to correlate changes in spectral features to changes in the structure of n-propylcobalamin on a subpicosecond timescale. These measurements provide a cobalt-centered movie of the excited molecule as it evolves to the local excited-state minimum.