OBSERVATION OF FEMTOSECOND, SUB-ANGSTROM MOLECULAR BOND RELAXATION USING LASER-INDUCED ELECTRON DIFFRACTION

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Imaging, or the determination of the atomic positions in molecules, has always occupied an essential role in physical, chemical and biological sciences. For structural determination, the well established methods of X-ray and electron diffraction easily achieve sub-Angstrom spatial resolution. However, these conventional approaches are not suitable for investigating structural transformations, such as the reaction of molecules or the function of biological systems that occur on the timescales faster than a picosecond. Over the past decade, major efforts directed at developing femtosecond pulsed sources, e.g. X-ray free-electron lasers and electron beams, have resulted in pioneering investigations on imaging large biological molecules and condensed phase dynamics. We report on a different approach, laser-induced electron diffraction (LIED), for achieving sub-femtosecond, sub-Angstrom spatio-temporal resolution for investigating gas-phase molecular dynamics. In contrast to the above mentioned techniques, the LIED method generates bursts of coherent electron wave packets directly from the molecule under interrogation. The study is performed by measuring the diffracted photoelectron momentum distribution produced by strong-field ionization of oxygen and nitrogen molecules at several mid-infrared wavelengths (1.7-2.3 μm). The bond lengths retrieved from the LIED analysis show sensitivity to a change of 0.05 Å in 1 fs. This initial report provides the first direct evidence of bond relaxation following an electronic excitation and establishes the foundation of the LIED method as a general approach for ultrafast imaging of molecular dynamics.