Abstract Submitted for the DAMOP15 Meeting of The American Physical Society

Probing ultrafast molecular dynamics in O_2 using XUV/IR pump-probe studies¹ D. RAY, F.P. STURM, T.W. WRIGHT, P. RANITOVIC, N.H. SHIVARAM, I. BOCHAROVA, A. BELKACEM, TH. WEBER, Chemical Sciences Division, Lawrence Berkeley National Laboratory — We investigate the molecular dynamics via different dissociative and autoionizing pathways in molecular oxygen using a pump-probe scheme with ultrashort extreme ultraviolet (XUV) laser pulses. Our primary focus is to study the molecular dynamics in the superexcited Rydberg states in a time-resolved manner. The O₂ molecules are pumped by 20.2 eV and 23.1 eV XUV pulses (13th and 15th harmonics). Probing the relaxation dynamics with an infrared (IR) pulse at very long delays (100s of fs) enables us to measure the lifetimes of these Rydberg states. We also observe an enhancement and suppression of vibrational levels of the O_2^+ ion due to the presence of IR. The high flux XUV pulses used for this experiment are generated in an Ar gas by IR pulses from our state-of-the-art 30 mJ, 50 Hz laser system. The pulses are overlapped with the supersonic jet in our Momentum Imaging for TimE Resolved Studies (MISTERS) setup. The cold target in our setup, combined with a very tight focussing geometry and a 3D momentum detection capability gives a high kinetic energy resolution. Molecular dynamics in other polyatomic molecules are also under investigation.

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Date submitted: 30 Jan 2015

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