Abstract Submitted for the DAMOP17 Meeting of The American Physical Society

Ultrafast Polarization Spectroscopy in Polyatomic Molecules RICHARD THURSTON, NIRANJAN SHIVARAM, ELIO CHAMPENOIS, SAID BAKHTI, PAVAN MUDDUKRISHNA, ALI BELKACEM, Chemical Sciences Division, Lawrence Berkeley National Laboratory — Polarization spectroscopy has been used in the past to study dynamics in solid, liquid and gas phase systems on picosecond and femtosecond time scales. In polarization spectroscopy, two laser pulses (drive and probe) with a relative polarization of 45 degrees, interact with the medium being probed. Due to the third order non-linear polarization induced in the medium a signal with a polarization orthogonal to the probe is generated along the probe direction. This signal measured after a crossed polarizer is directly related to the induced birefringence and dichroism in the medium. Here, we present preliminary measurements in ultraviolet grade fused silica and discuss our model to obtain the ultrafast electronic response of the medium. We then discuss the extension of this method to study ultrafast dynamics in polyatomic molecular systems using multiple pulses.

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Date submitted: 02 Feb 2017 Electronic form version 1.4