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Ultrafast Transient Polarization Spectroscopy of Electronically Excited Molecular Systems[1] RICHARD THURSTON, NIRANJAN SHIVARAM, ELIO CHAMPENOIS, SAID BAKHTI, PAVAN MUDDAKRISHNA, ALI BELKACEM, DANIEL SLAUGHTER, 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. 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. By introducing in-phase and out-of-phase local oscillators in conjunction with lock-in amplification, the signal after a cross polarizer gives a sensitive measurement of the real and imaginary components of the medium's third order non-linear response. Here, we discuss this method as applied to electronically excited systems and present preliminary measurements of the ultrafast electronic response of liquid nitrobenzene. We then extend this method to the study of ultrafast dynamics in gas phase polyatomic molecular systems and discuss the enhanced sensitivity of this method compared to ultrafast transient absorption spectroscopy.

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