Abstract Submitted
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Ultrafast Dynamics in Molecular Systems Studied Using Polarization Spectroscopy[1] NIRANJAN SHIVARAM, ELIO CHAMPENOIS, SAID BAKHTI, RICHARD THURSTON, PAVAN MUDDUKRISHNA, ALI BELKACEM, Chemical Sciences Division, Lawrence Berkeley National Laboratory — Studying ultrafast dynamics in complex polyatomic molecules using photo-electron and photo-ion spectroscopy can be very challenging due to the presence of many competing processes. Photon-in photon-out methods like the widely used transient absorption spectroscopy can be very helpful in isolating some of these processes. Here, we discuss a different approach to measure ultrafast dynamics in molecules using the technique of polarization spectroscopy which is a special case of four-wave mixing. Two pulses (drive and probe) with a relative polarization of 45 degrees interact via the third order susceptibility of a medium. The signal generated along the probe direction with a polarization orthogonal to the input probe polarization is measured using a crossed polarizer. We first demonstrate the method using two weak near infra-red (IR) femtosecond pulses from a Titanium Sapphire laser system to study the electronic response in ultraviolet (UV) grade fused silica. We then discuss the extension of this method to study ultrafast excited state dynamics in molecules like O-nitrophenol using UV and two IR pulses. This method has the potential to be more sensitive than transient absorption by 2 to 3 orders of magnitude without the requirement of resonant transitions to probe the dynamics.

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