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Ultrafast transient polarization spectroscopy as a sensitive probe of photon-driven excited state dynamics of nitrobenzene.[1]

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I will discuss a novel approach to measure ultrafast processes in molecules based on the technique of Optical Kerr-effect (OKE) spectroscopy. In conventional OKE spectroscopy, 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, resulting in a near-background free, highly sensitive measurement. In our adaptation of the method, we use an additional pulse to first pump the system to an excited state and use the drive/probe pulse-pair to probe the dynamics that follow. We demonstrate this method in liquid Nitrobenzene using a femtosecond ultraviolet pulse as pump and near infrared pulses as drive and probe. We perform this measurement in an optical heterodyne configuration which enhances signal levels in addition to allowing a direct extraction of the non-linear response of the medium. This method has the potential to be more sensitive than transient absorption by about three orders of magnitude without the requirement of resonant transitions to probe the dynamics.

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