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Early optical response of fluorescent molecules studied by synthetic optically delayed pulses[1] ARKAPRABHA KONAR, JAY SHAH, TAPAS GOSWAMI, VADIM LOZOVOY, MARCOS DANTUS, Michigan State University — The early optical response of fluorescent molecules in solution is probed by pairs of collinear pulse replicas. Two approaches are followed. First approach mimics an interferometer, replicating interference as a function of time delay between the pulses. For the second approach, each pulse spans the entire laser bandwidth, sharing no common frequencies with the second pulse, thus no interference is observed between the pulses. In both cases, the pair of pulses is delayed with attosecond resolution to study IR 144. Both fluorescence at right angles or the stimulated emission along the output beam as a function of time delay is monitored. At high intensities when approximately 10% of the dye molecules are excited, the second pulse can stimulate emission from molecules excited by the first pulse, thereby giving rise to interference fringes every 2.66 fs. When the pulse replicas are generated by multiple independent comb shaping, it is evident that the interference fringes for stimulated emission bear an out of phase relationship with those observed from fluorescence and have a maxima at time zero. This is masked with conventional pulse replicas that interfere.

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