Multimodal Fieldoscopy

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Non-invasive, label-free vibrational spectroscopy characterizes complex molecular compositions by leveraging unique molecular resonances for inherent chemical selectivity. Stimulated Raman spectroscopy (SRS) and short-wavelength infrared (SWIR) absorption spectroscopy complement each other with low water absorption, deep penetration, and linear molecular concentration dependence. We present broadband, field-resolved detection of stimulated Raman gain and SWIR absorption in liquids with attosecond resolution. Our technique enables background-free, simultaneous detection of SWIR absorption and the SRS fingerprint region, capturing both amplitude and phase information.

The experimental setup is shown in figure 1a. 40 µJ, 250 fs pulses at 1030 nm and a 1 MHz repetition rate are equally split into two paths. In one path, the pulses are frequency doubled and used as a pump for optical parametric amplification (OPA). In the other path, a supercontinuum with a pulse duration of 4.5 fs is generated in two stages of gas filled hollow-core fibers. 4% of these pulses are reflected via a beam splitter into a delay line and used as gate pulses for field detection. The 96% transmitted pulses are generating broadband CEP stable 15 fs Stokes pulses with a spectrum from 1.5 µm to 2.6 µm via intrapulse difference frequency generation (IPDFG). An etalon is used to filter out a 3 ps Raman pump at 1375 nm from the remaining supercontinuum from the IPDFG stage. The Raman pump is modulated at 500 kHz via an acousto optical modulator for lock-in detection and is amplified via an OPA up to 300 mW. The Raman pump is collinearly combined with the Stokes and focused into liquid flowing in a 400 µm thick capillary. In the liquid, energy is transferred from the Raman pump to the Stokes resulting in Raman gain as well as a broadband Kerr gain amplifying the entire electric field of the Stokes. After the sample, the Raman pump is filtered out and the Stokes is combined with the gate pulses measuring the gain in the Stokes' electric field via electro optic detection. Figure 1b depicts the measured electric field of the stokes pulses in liquid acetonitrile. Figure 1c shows the corresponding Fourier transformation of the molecular response in the temporal window of 200 fs up to 2100 fs. The Raman features of acetonitrile in the fingerprint region as well as linear SWIR absorptions are visible demonstrating multimodal fieldoscopy.

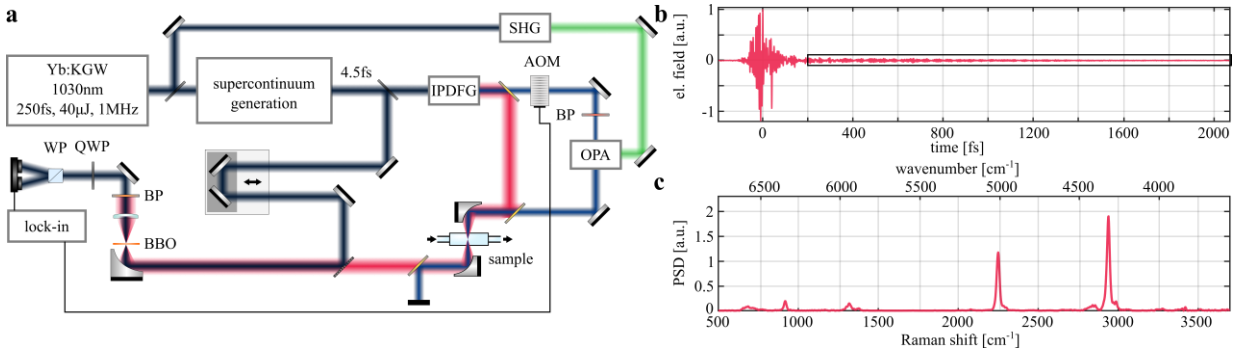

Figure 1. a) Experimental setup of multimodal fieldoscopy. b) Field-resolved response of liquid acetonitrile. c) Fourier-transformation of the molecular response showing Raman peaks as well as linear SWIR absorption peaks